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Rheological Rheological Characterization of Bitumen/Waste Polyethylene Composites: Part II

Mahmoud Abdel-Halim Abdel-Goad

Chemical Engineering Dept., Faculty of Engineering, Minia University, Egypt
E-mail:m.abdelhalim@mu.edu.eg

Abstract: *Bitumen blends were prepared for road applications by the introduction of 9wt% waste Polyethylene, PE bags in the molten state. The relaxation stress, relaxation and retardation spectrum and viscosity of bitumen blends were studied. At different temperatures and compared to those of the original pure bitumen. These properties were tested using an ARES- Rheometer (Rheometric Scientific, Co.) equipment. The measurements were performed in the dynamic mode, plate-plate geometry of 8 mm diameter over the temperature range from –10 to 60°C and angular frequency, ω varied from 10^{-1} to 10^{2} radian/s. The relaxation stress and viscosity were modified by the addition of waste PE. The results also evidence that the incorporation of the waste PE enhances stability of the bitumen blends As a consequence, the use of waste PE bags can be considered a suitable alternative from both environmental and economical points of view. Bitumens containing several polymer concentrations have been studied.*

Keywords: bitumen, waste plastics, relaxation stress, relaxation spectrum, viscosity.

1. INTRODUCTION

Bitumen, which remains after vacuum distillation of crude oil, is a complex mixture of organic and inorganic compounds. The physical, mechanical, and rheological behavior of bitumen in road and building construction is governed by its chemical composition. For this reason, considerable effort goes toward widening the knowledge of this composition and relating it to performance [1-8].The chemical composition of bitumen is very complex, although its components can be broadly categorized as maltenes and asphaltenes.[9- 14]. Asphaltenes are defined as the black-coloured fraction of bitumen that is insoluble in *n*-heptane. Maltenes, composed of saturated compounds, aromatic compounds and resins are soluble in *n*-heptane . The ratio of asphaltenes to the other constituents, and the maltenic fraction composition have a significant effect on the viscoelastic properties of bitumens, and hence on their performance as road paving binders [15].

The high temperature sensitivity makes bitument difficult or impossible to optimise both high- and low-temperature properties. The binders are too brittle at low temperatures and lack cohesion at high temperatures. The temperature sensitivity of straight-run bitumens can be reduced by adding polymer [16].

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 (K_{1C}) at low temperature of polymer modified bitumen were shown to be higher than those of the bitumen base [17]. Therefore, polymer-bitumen blends find considerable interest from different authors[18- 21].

The application of polymeric materials in agriculture and horticulture has increased considerably in recent years, not only as replacement for traditional materials but also as a mean of effecting improvement. Thus, the application of plastics has led to a significant improvement in technological processes in the growing and storing of agricultural crops . As a result, a large amount of waste plastic films, containing LDPE, EVA and a minor proportion of stabilizers, are generated every year all over the world [22]. Therefore, the possibility of disposing of troublesome waste plastics within road bitumens is an environmental and economical point of view,

The rheological analysis of the bituminous materials is an important test to better understanding its microstructure. Therefore, the rheological study has a considerable interest by different authors [23-27]. It is well established that the linear behavior of bitumens is described by the general linear visco-elastic model.The rheological analysis is a sensitive and versatile thermal analysis technique, which measures the modulus (stiffness) and damping properties (energy dissipation) of materials as the material are deformed under periodic stress [28].

Since the modulus of bitumen can vary by 3–4 orders of magnitude over the in-service temperature range. It was the objective of this work to modify the bitumen at in-service temperatures by adding waste PE. we analyze the rheology of the bitumen modified with waste PE content up to 11% and neat bitumen.

2. EXPERIMENTS PART

Materials and preparation

Bitumen blends were prepared by adding waste plastics to commercial bitumen. The waste PE bags were sorted from the garbage and shredded into coarse particles. The waste PE bags and the bitumen were weighted and heated individually in an oven until melt. Waste PE in molten state was poured into the molten bitumen up to a level of 11 wt%, mechanically stirred to give a homogenous sample and then stored at room temperature. The bitumen blends and the original neat bitumen samples were compression-molded at 70°C and 5 bars for 1 hour and shaped for the rheology measurements in a disc form with 8 mm diameter and 1 mm thickness.

3. METHOD

The rheological behavior was studied for pure bitumen and bitumen blends by using dynamic an ARES-Rheometer (Rheometric scientific) equipment in plate-plate geometry of 25 mm diameter. The sample to be measured was placed on the down plate and the upper plate was lowered until just contact the sample. The measurements were performed in the temperature range from –10 to 60°C and angular frequency, ω varied from 10^2 to10^{-1} radian/s. In the dynamic mode and the strain amplitude was kept (1%) in the linear viscoelastic regime. 6 points per decade in frequency were obtained. The gap was 2 mm and the actual gap size is read electronically and allows absolute moduli to be determined.

4. RESULTS AND DISCUSSIONS

The master curve is obtained by shifting the measured data at different temperatures, T into a single curve at a reference temperature T_0 as shown in Figure 1. By using of the time-temperature superposition principle which described by Williams-Landel-Ferry [29] as, $\log a_T = -C_1 (T-T_0) /(C_2+(T-T_0))$.Where a_T is the horizontal shift factor and constants C_1 and C_2 are material specific. C_1 and C_2 are roughly determined as listed in Table1. a_T of bitumen blends is plotted as a function of temperature in Figure 2. In this Figure a_T decreases with increasing the temperature up to 25°C ($a_T \sim T^{-0.14}$) then becomes nearly independent on the temperatures. a_T shifts the data obtained at different temperatures in the horizontal direction. In vertical direction the experiments data are shifted by b_T ($b_T = \rho T / \rho_0 T_0$ [30]). Where ρ is the material density. The effect of the temperature on b_T is shown in Figure 3 for bitumen blends. b_T is nearly constant at low temperatures but after 25°C it falls rapidly ($b_T \sim T^{-0.17}$) with increasing the temperature as shown in Figure 3.

Material	C_1	C_2
Bitumen base	24.9	196
Bitumen/9%PE	1.8	30

The shear relaxation stresses G(t) is plotted against time with logarithmic scales at T_0 = 10, 25 and 60°C in Figures 4-5 for neat bitumen and bitumen/PE blend. G(t) is the relaxation stress at constant deformation. At long times of each T_0, the master curve of each G(t) shows flow regime which is related to the complete relaxation of the macromolecules and released from the restrictions of the entanglements. The time taken in the relaxation processes is called the longest relaxation time. In this regime the viscous component is higher than the elastic component because of the release of the entanglements which are the responsible for the elasticity. Therefore, G(t) decreases sharply and falls by many order of magnitude in this regime as shown in Figures 4-5. At intermediate time of each master curve is located the rubbery zone that is the signature of the elasticity. As a result, G(t) is independent on the time in this regime since the loss in the energy in every cycle of the deformation is minimum. At short times the third regime of the viscoelastic behaviour is found only in the case of pure bitumen which is called dynamic glass transition because it was no difficulties to measure the neat bitumen samples at very low temperatures (-10°C). The master curves at reference temperature 10°C are shifted toward the long time, those at 25°C appear to be in the intermediate and the master curves at reference temperatures 60°C are shifted toward the short time-wing as shown in Figure 4. This because the time of relaxation of the entanglement chains is inversely proportional to the temperature, since at low temperatures the chains take a long time in the relaxation processes, unlike at high temperatures. G(t) modulus at 60°C is lower than at 25°C and G(t) at 25°C is lower than at 10°C as seen in Figures 4-5.

Figures 4-5 evidence that the incorporation of the waste plastics into the bitumen increases the relaxation stress over all the temperatures and times ranges. This increase in G(t) moduli is more noticeable at long times and high temperatures as shown in these Figures. Where the values of G(t) modulus at 5 seconds and 10°C is 2 times higher than this of neat bitumen by by the addition of 9wt%PE as shown in Figure 4. This factor reaches up to 7 at 50 seconds (the same temperature) as shown in Figure 4. The ratio $G(t)_{bit.-9\%PE}/ G(t)_{bitumen}$ at the end of flow regime in the cases of T_0 = 10, 25 and 60°C are 3, 6 and 10. That means the bitumen/PE blend is more stable than neat bitumen at high temperatures for a long times. This due to the formation of the bitumen-polymer network which increases the stiffness and strength of the bitumen blends. That is confirmed too in Figures 6-7. In these Figures the shear equilibrium creep, Je is logarithamically plotted versus time for bitumen and bitumen/PE blend. The addition of PE to bitumen enhances Je in particular at long times and the differences among Je values of bitumen/PE blend is more observable at long times due to the stability of the bitumen-PE network.

The relaxation (H(t)) and retardation (L(t)) spectrum of neat bitumen and bitumen/PE blend are plotted as a function of time at 25°C in Figures 8-9. H(t) and L(t) indicate the distribution of relaxation and retardation mechanisms, respectively in different regions of the time scale. they show a broad spectrum of relaxation and retardation times. A broad mechanical relaxation, observed at 25°C, is associated with the collapse of a compact structure, constituted by asphaltene particles surrounded by solid resin. The addition of PE to bitumen modifies this relaxation.

The values of H(t) increase by the addition of waste PE to bitumen. The increase in H(t) is significant at long times as shown in Figure 8. That is clear at 40 seconds where H(t) of bitumen blend is higher than this of pure bitumen by a factor 2 by the addition of 9% PE as shown in Figure 8. This factor becomes 6 at 100 seconds as shown in Figure 8. The same scenario is observed in L(t) (Fig.6). Since at 40 Seconds the addition of 9% PE to bitumen alters the values of L(t) for pure bitumen from 5.0×10^{-5} to 4.3×10^{-1} Pa^{-1} as shown in Figure 9. But at 1000 seconds the values of L(t) are 5.0×10^{-3} and 5.7×10^{-4} Pa^{-1} for pure bitumen and 9%PE , respectively as shown in Figure 9. This as explained earlier due to the increase in the stiffness and strength of the bitumen by the incorporation of the PE.

The complex viscosity (η*) as a function of temperature for neat bitumen and bitumen blends at ω = 0.1 radian/s is plotted in Figure 10. As shown in this Figure the incorporation of the waste PE into the bitumen increases the viscosity over all the temperatures range. This because the viscosity of the bitumen matrix is higher than pure bitumen. η* of the blends at high temperatures are higher than at low temperatures. Since the values of η* at 15°C increase from 5.0×10^{4} (pure bitumen) to 5.7×10^{5} Pa.s by the addition 9wt% PE , respectively. But at 60°C the differences among the values of η* are high. These values are 2.7×10^{5} and 5.7×10^{5} Pa.s for pure bitumen and bitumen-9%PE, respectively as shown in Figure 10. This difference because bitumen components change considerably at ~ 40°C, particularly the ratio of the asphaltenes to the maltenes (the main components of bitumen) which has a significant effect on the viscoelastic properties of bitumen.

5. CONCLUSION

Bitumen blends were prepared as a road paving binders by the introduction of 9wt% waste PE bags in the melt state followed by mechanical stirring. The relaxation stress, relaxation and retardation spectrum and viscosity of bitumen blends were studied. At different temperatures and compared to those of the original pure bitumen. These properties were tested using an ARES-Rheometer (Rheometric Scientific, Co.) equipment. The measurements were performed in the dynamic mode, plate-plate geometry of 25 mm diameter over the temperature range from -10 to 60°C and angular frequency, ω varied from 10^{2} to10^{-1} radian/s. The relaxation stress and viscosity were developed by the addition of waste plastics and increase with the waste plastics content. The results also evidence that the incorporation of the waste plastics enhances stability of the bitumen blends. The results showed an increase in these properties by the incorporation of the bitumen at high temperatures for a long time.

6. ACKNOWLEDGMENTS

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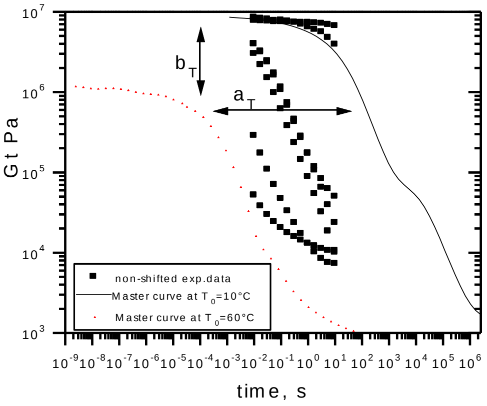

Figure 1: non shifted experiments data of G(t) at different temperatures are shifted into master curves at T_0 =25°C by using a_T and b_T.

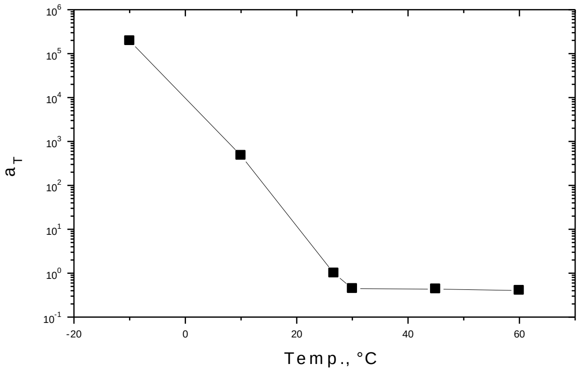

Figure 2: a_T of bitumen/PE blend as a function of temperature at T_0 =25°C

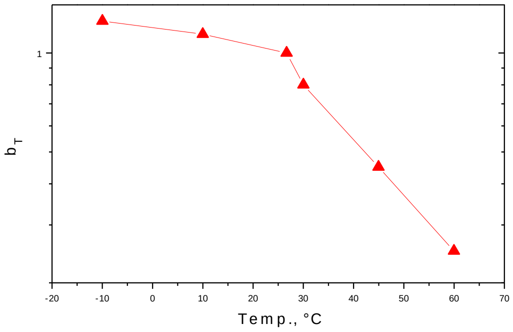

Figure 3: b_T of bitumen/PE blend as a function of temperature at T_0 =25°C

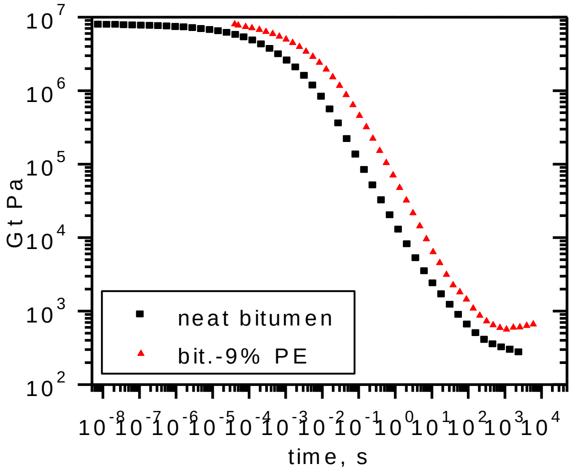

Figure 4: G(t) of bitumen/PE and neat bitumen at T_0 = 25°C

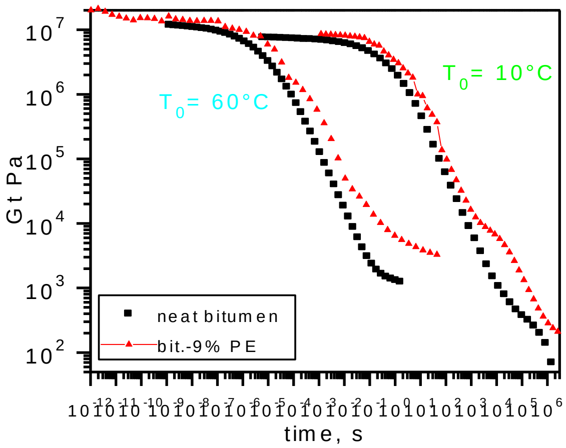

Figure 5: G(t) of bitumen/PE and neat bitumen at T_0 = 10 and 60°C

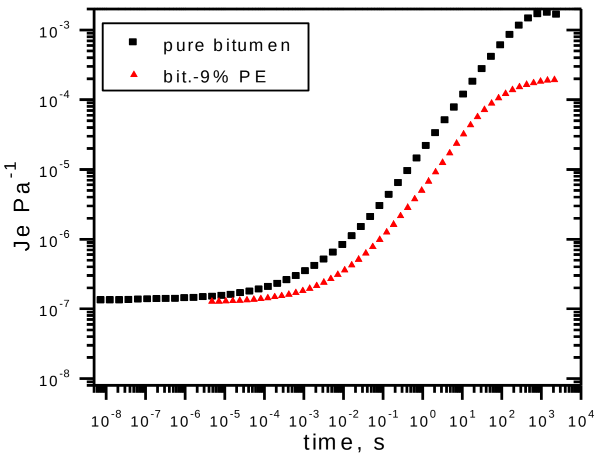

Figure 6: Je of bitumen/PE and neat bitumen at T_0 = 25°C

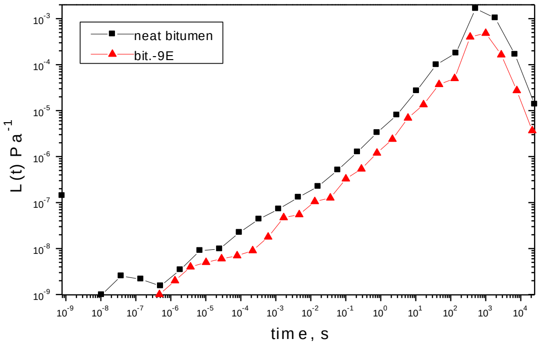

Figure 9: L(t) of bitumen/PE and neat bitumen at T_0 = 25°C

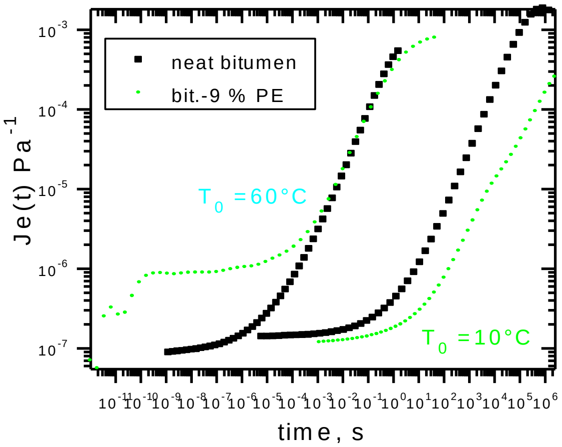

Figure 7: Je of bitumen/PE and neat bitumen at T_0 = 10 and 60°C

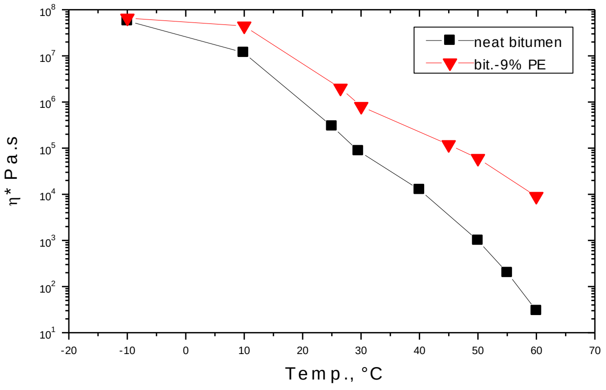

Figure 10: η* of bitumen/PE and neat bitumen as a function of temperature at ω = 0.1 rad/s

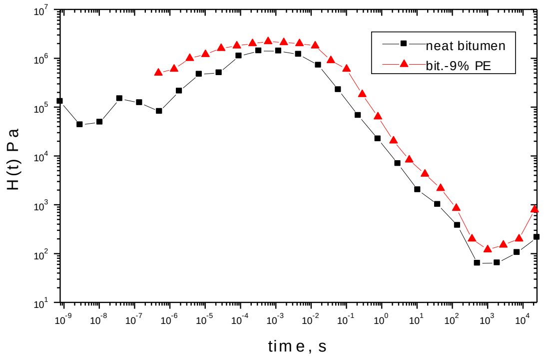

Figure 8: H(t) of bitumen/PE and neat bitumen at T_0 = 25°C