

Effect of plastic bag waste addition on the rheology properties of a bitumen

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ARTICLE INFO ABSTRACT/RESUME Article History: Abstract: Statistical figures indicate that the 7 billion of plastic bags, thrown annually in the nature in Algeria represent a real threat to the Received : 02/02/2020 environment and call for urgent action measures. Several studies have Accepted : 28/09/2020 shown the potential of waste plastic recycling in producing high performance road paving bituminous materials. The performance of Key Words: road paving asphalt mixtures depends largely on the rheological Bitumen; properties of the bitumen used to bind the aggregate. This contribution Modified bitumen; studies the effects of plastic bag waste addition on the rheological Plastic bags waste; properties of a waste plastic modified bitumen (WPmB). Basic tests Rutting resistance and dynamic rheological tests were used to characterize the modified binder. FTIR spectroscopy was also called upon to disclose the chemical functionalities present in the waste plastic modified bitumen (WPmB). Results obtained show that interesting improvements are observed on the rheological properties of the WPmB. It was found that plastic addition stiffens the bitumen. The dynamic rheological results show that the plastic addition increases the complex modulus and consequently the complex viscosity and decreases the phase angle of the WPmB at all temperatures and frequencies tested. Morwever, plastic addition improves the rutting resistance of WPmB as defined by the SHRP criterion.

I. Introduction

8.3 billion tons of plastic waste have been produced between 1950 and 2015 in the world. 79% of this plastic waste ended up thrown in the nature, 12% were incinerated and only around 9% have been recycled [1]. Geyer et al. [1] showed that if current production and waste management trends continue, roughly 12,000 Mt of plastic waste will be thrown in the environment or in landfills by year 2050. France is the biggest producer of plastic waste in the Mediterranean area (with 4.5 million tons/year; 66.6 kilos/person) from which 22% are recycled, 40% incinerated, 36% buried and 2% are thrown in the nature [2]. In Algeria, the industrial waste management system is still not enough developed technically and organizationally to cope with the real waste problem of the country. Plastic waste represent a significant fraction of municipal solid waste; more than 1.2 million tons per year; about 12% (Figure 01) [3]. According to the figures of the National Agency of Waste [4], the percentage of plastic waste grew to 17% of municipal solid waste in 2018. Algeria is ranked fifth country in the world in plastic bag utilization with about 7 billion plastic bags annually [5]. Only 10% of these has been recovered in various fields [4], the rest is thrown in the nature representing a real threat to the environment.

The Algerian road construction industry is experiencing the introduction of new highperformance materials such as the so-called high modulus asphalt (Enrobé à Module Elevé) in order to meet the ever increasing mechanical requirements of road pavements. The EME material is normally manufactured using a local aggregate, a conventional bitumen, and a chemical additive. The chemical proprietary additive, is imported from abroad. It is manufactured in the form of granules and incorporates some sort of recycled waste plastic as a main component. Local experience has shown that manufactured chemical additives can effectively improve the performance of the asphalt mix giving high modulus asphalt materials (EME), but leaves the plastic bag waste problem of the country entirely posed.

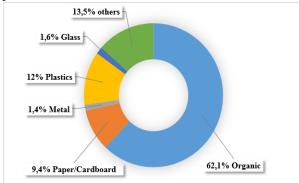


Figure 01. Municipal solid waste composition in Algeria [4].

Several investigations were performed on the reuse of plastic waste in improving road materials. Two methods were used to incorporate waste plastic into road pavement materials, the wet method that incorporates the waste plastic into the bitumen obtaining so doing a modified bitumen, and the dry method that adds directly the waste plastic to the aggregates and bitumen of the asphalt mix.

As for the dry method, results have shown that, plastic waste addition improves significantly the physical and the mechanical characteristics of the asphalt mix such as the Marshall stability, the Marshall quotient, the indirect tensile strength, the fracture energy, the resilient modulus, the permanent deformation and the creep compliance of the asphalt mix [6]–[10]. Waste plastic LDPE carrying bag modified asphalt mix shows a better binding property, stability, density, fatigue life, and moisture susceptibility relatively to the control mix

[11]–[14]. Rokade [15] used both dry method for waste plastic LDPE modified asphalt mix (LDPEmA) and wet method for Crumb Rubber modified bitumen (CRmB) on the same type of mixture. The results reveal that the Marshal stability value increased by about 25 % and the density of the asphalt mix increased with LDPEmA and CRmB addition. In the dry process (LDPEmA), plastic waste is coated over aggregate which helps to have a better binding of bitumen with the plastic-waste coated aggregate due to increased bonding between plastic and bitumen.

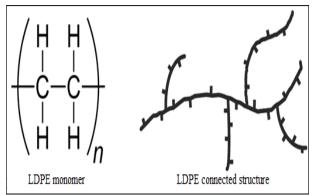
As for the wet method, Costa et al. [16] demonstrated that it is possible to obtain similar properties, or even better, than those of commercial polymer modified bitumen when using the waste plastic as bitumen modifiers. The waste plastic (LDPE) modified bitumen shows more profound effect on penetration rather than softening point [17]-[19]. The viscoelastic behavior of plastic modified bitumen depends on the plastic waste content, mixing temperature, mixing technique, and also on molecular structure of plastic polymer used [20]. Results have shown that recycled LDPE modified bitumen leads to an increase in the values of the storage and loss moduli, and viscosity, as well as an apparent decrease in thermal susceptibility and phase angle compared to the original binder [20]-[22]. Moreover. the mechanical glass transition temperature is reduced and, consequently, the glassy region is shifted to lower temperatures [21]. Zhang et al. [23] and Farahani et al. [24] have shown that no chemical reactions take place between the waste plastic and the bitumen components indicating that the bitumen modification with waste plastics is a purly physical process.

This study address the characterization of the rheological properties of plastic bag waste modified bitumen. A waste plastic modified binder is characterized by DSR (Dynamic Shear Rheometer) testing to assess its viscoelastic behavior at medium and high temperatures. Thermal (DSC, TG) and FTIR analyses are used to complete the characterization of the plastic modified binder.

II. Materials and methods

The base bitumen used in this study is a 50/70 penetration grade. It is supplied by the L2MGC laboratory (Cergy Pontoise University). Its penetration and softening point values were measured to be respectively 52.5 dmm and 50 °C. The waste plastic bags used are goods carrier bags collected from a local market. Generally, the plastic bags are made of Low-Density Polyethylene (LDPE). The LDPE polymer is characterized by a connected structure (Figure 2). Its chemical formula is $(C_2H_4)n$, as shown in Figure 2. The LDPE density varies between 0.915-0.935 and its melting temperature is measured to be around 130 °C as shown by the DSC results (Figure 12). In order to be perfectly incorporated into the bitumen, the waste plastic was shredded down into small particles of average size 2-5 mm (Figure 3).





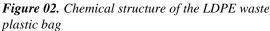




Figure 03. Shredded waste plastic bag of average size 2-5 mm

II.1. Preparation of modified bitumen

The waste plastic particles are progressively added to and mixed into the previously heated pure bitumen at 170 °C with a 1200 rpm speed stirring device for a period of 60 min using Heidolph RzR 2041 agitator. The waste plastic content has been fixed at 0.7% by weight for this study.

II.2. Test methods

Penetration and softening point temperature were measured according to the EN 1426 and 1427 standards respectively. The rheological properties of the pure and modified bitumen samples were measured in the linear viscoelastic range using a controlled stress rheometer (Haake RT 20) with a profiled parallel-plate geometries 20 mm diameter and 1 mm gap. The dynamic viscoelastic properties, at in-service and high temperatures, were studied for temperatures ranging from 20 to 80 °C for a frequency sweep 0.1 - 10 Hz range. The FTIR spectra of the plastic bag waste (WP), pure bitumen (PB) and WPmB have been measured on the Bruker Tensor 27 spectrometer following the ATR procedure. Thermal transitions of materials when heated have been measured by the differential scanning calorimetry (DSC) in a nitrogen atmosphere using a NETZSCH apparatus for a temperature going from 20 to 200 °C with a heating rate of 5 °C/min.

III. Results and discussion

III.1. Penetration and softening point of WPmB

As can be seen from the results of Figure 4, the waste plastic bag addition decreases the penetration value from 52 for the PB to about 37 (dmm) for the 0.7WPmB and the softening point temperature increases from 50 for the PB to about 56 °C for the 0.7WPmB. As shown by the literature the plastic bag waste has a greater effect on the penetration rather than on the softening point [17]–[19]. The results obtained indicate that the waste plastic addition can improve substantially the bitumen high temperature resistance and consequently the asphalt mix performance using the WPmB as a mix binder.

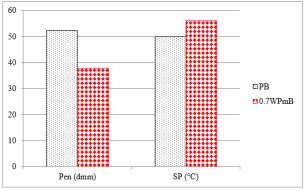


Figure 4. Penetration and softening point of PB and WPmB

III.2. Dynamic rheological results of PB and 0.7WPmB

Curves of isothermal complex modulus, phase angle and complex viscosity of PB and WPmB at 20, 50 and 80 $^\circ C$ are shown in Figure 5, 6 and 7 respectively. Both PB and 0.7WPmB show a similar qualitative behavior characterized by a continuous increase in the viscoelastic moduli and a continuous decrease in the Tan φ and the complex viscosity with frequency. It can be seen that waste plastic addition increases both the complex modulus and the complex viscosity and decreases the Tan φ of the modified bitumen even though the observed increase in both complex modulus and viscosity is low for ambient temperature and is more substantial at higher temperatures. The substantial increase of the complex modulus and the viscosity, at 50 and 80 °C, may improve the rutting resistance of the asphalt pavement using the WPmB as a binder [25]. It can be deduced from the results of the phase angle that the plastic addition improves the elastic properties and reduces the viscous behavior of the modified bitumen especially at high temperatures.

This better resistance to high temperatures of the WPmB can stem from the high melting temperature (around 130° C) of the waste plastic bag relatively to that of the pure bitumen. The same effect has been observed in the increase of the softening point temperature of the WPmB (see Figure 4).

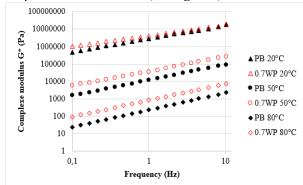


Figure 5. Isothermal of complex modulus of PB and WPmB at 20, 50 and 80 °C

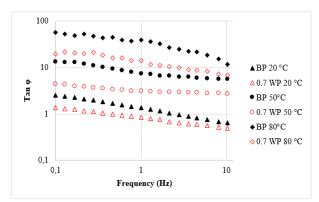


Figure 5. Isothermal of phase angle of PB and WPmB at 20, 50 and 80 °C

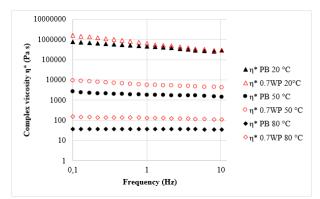


Figure 7. Isothermal of complex viscosity of PB and WPmB at 20, 50 and 80 °C

III.3. Black and Cole-Cole diagrams

Black and Cole-Cole diagrams of the PB and the WPmB are shown in Figure 8 and 9 respectively. The experimental points of the two curves for both PB and WPmB are continuous and unique indicating that the two binders undergo the time-temperature superposition principle in the linear viscoelastic

range. The Black diagram results (Figure 8) indicate that there is a monotonic decrease of the phase angle with respect to the complex modulus. The effect of the waste plastic bag addition on the rheological behavior of the bitumen is evidenced on the Black diagram by the increase of the complex modulus and the decrease of the phase angle at a given temperature or frequency compared to the pure bitumen. The Cole-Cole diagram (Figure 9) shows a linear increase of G" versus G' at medium and high temperatures. As the low temperatures have not been tested in this work, the shape of the Cole-Cole diagram is not parabolic as normally expected. The experimental points of the two curves show that the waste plastic addition improves the bitumen elastic behavior explained by the move of the curve towards the more elastic part.

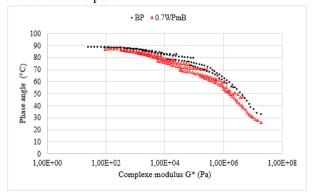


Figure 8. Black diagram of PB and WPmB

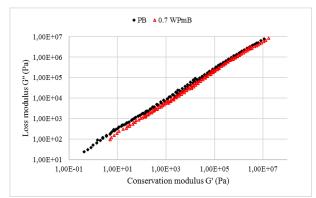


Figure 9. Cole-Cole diagram of PB and WPmB

III.4. Rutting resistance of PB and WPmB

In accordance with the SHRP specifications, the rut factor G*/sin φ at 1.6 Hz (10 rad/s) was used to show the effect of the waste plastic bag addition on the improvement of the rutting resistance of the modified bitumen. Results of Figure 10 show that the 0.7WPmB rut factor is greater than that of the PB over the whole temperature range. According to the SHRP criterion, the temperature causing the rutting corresponds to the temperature at which the parameter G*/sin φ = 1000 Pa. It can be seen that the addition of only a small amount of waste plastic increases significantly the rutting temperature by about 10 °C compared to the pure bitumen.

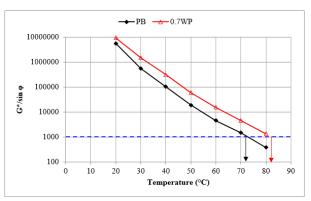


Figure 10. rut factor G^* / sin φ at 1.6 Hz of PB and **WPmB**

III.5. FTIR spectra analysis of WP, PB and **WPmB**

The FTIR spectra of the plastic bag waste, the pure bitumen, and the 0.7WPmB are plotted in Figure 11. The PB and the WPmB show the same spectral bands. The absorption peaks at 2918 and 2850 cm⁻¹ correspond to the asymmetric and symmetric stretching vibrations of C-H in methylene -CH2respectively. The peaks at 1455 cm⁻¹ correspond to the scissoring vibration of methylene -CH2-. The peaks at 1376 cm⁻¹ correspond to the umbrella vibration of methyl -- CH3. Spectral bands at 809 cm-1 correspond to the stretching vibrations of benzene and the peaks at 720 cm⁻¹ correspond to the vibration of $(-CH_2-)n$, $n \ge 4$ [26], [27]. According to the results, WPmB undergoes no observable changes in its functional groups relatively to the PB indicating that there is evidently no chemical reaction that takes place between the plastic of bags and the bitumen components. The modification that entails the bitumen by addition of waste plastic is of a physical nature [28].

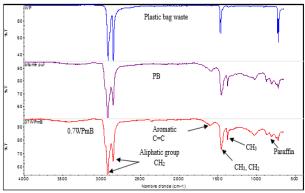


Figure 11. FTIR spectra of WP, PB and WPmB

III.6. Differential Scanning Calorimetry DSC of WP, PB and WPmB

The ATG and DSC spectra of the plastic waste is plotted in Figure 12 and those of PB and WPmB in Figure 13. The gravimetric thermal curve of the plastic shows that there is no mass loss in the WP up to 200 °C indicating that the decomposition temperature of plastic is greater than 200 °C. The DSC results show that the melting temperature of WP is around 130 °C. As for the WPmB, the endothermic peak which appears at 122 °C, is associated to the melting of the plastic of the modified bitumen. The shifting of the melting temperature from 130 °C (pure plastic) to 122 °C (WPmB) may indicate the physical interaction that takes place between the plastic particles and the oily fraction of the pure bitumen (following the plastic particles swelling by the aromatic oil). The results show also that the waste plastic bag addition increases the temperature at which the bitumen state changes from elastic solid to viscous fluid, from about 50 °C for the pure bitumen to about 70 °C for WPmB indicating a better thermal behavior of the WPmB over that of the pure bitumen. However, no exothermic peak was observed in the WPmB indicating that there is no chemical changes that occurs in the mixture, which confirms the results obtained by the FTIR spectroscopy presented above (Figure 11).

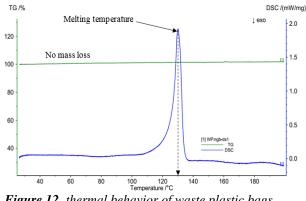


Figure 12. thermal behavior of waste plastic bags

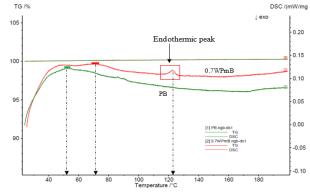


Figure 13. Thermal behavior of pure bitumen and waste plastic bags modified bitumen

IV. Conclusion

The results obtained in this study show that the waste bag plastic, predominantly composed of LDPE, addition improves significantly the physical and rheological properties of the base bitumen. In effect, the penetration value decreases and the softening point temperature increases with plastic addition. The dynamic rheological results indicate that waste plastic addition increases substantially both complex modulus and consequently the complex viscosity at high temperatures. Plastic addition improves the elastic properties and reduces the viscous behavior of the modified bitumen especially at high temperatures.

Black and Cole-Cole diagrams show that the timetemperature superposition principle in the linear viscoelastic behavior apply for both PB and WPmB. The effect of the waste plastic addition on the rheological behavior of the bitumen is evidenced on the Black diagram. In accordance with the SHRP specifications, the addition of only a small amount of plastic bag waste increases significantly the rutting temperature of the WPmB by about 10 °C compared to the pure bitumen as indicated by the rut factor $G^*/\sin\varphi$ at 1.6 Hz. There is no chemical reaction that takes place between the plastic of bags and the bitumen components as indicated by FTIR spectra. The DSC results indicate that the WPmB present a better thermal behavior comparatively to the pure bitumen. The results are promising provided that the local plastic bag waste can improve substantially the engineering properties of the Algerian road pavements. Future work will address the application of the bitumen/plastic blend on real road sections in order to demonstrate the superior performance of the pavement realized with WPmB under real traffic and climate conditions.

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