VALUE-ADDED APPLICATION OF WASTE PLASTIC ADDITIVES IN RECLAIMED ASPHALT PAVEMENT MIXTURES- RECYCLING WASTE AS SUSTAINABLE PAVEMENT MATERIALS

ABSTRACT

Waste plastic is a major category of municipal solid waste in Hong Kong and presents a significant challenge in disposal. This study develops an innovative approach to use waste plastic additives in pavement mixtures containing reclaimed asphalt pavement (RAP), which is also a waste material classified as construction waste. In recent years, the use of RAP for road pavement construction has been widely encouraged due to environmental and economic benefits. However, the percentage of RAP is usually capped at 20% to 25% as studies have shown that a high percentage of RAP might be detrimental to overall pavement performance. Research has shown that the addition of waste plastic materials such as polyethylene terephthalate (PET) into asphalt pavement may potentially improve the durability and overall performance of pavement mixtures. In this paper, the feasibility of using waste PET derived additives to improve the performance of hot mix asphalt (HMA) mixtures containing a high percentage of RAP binder is evaluated. To achieve this objective, various binder samples were prepared. These samples were characterised through various laboratory tests, including moisture susceptibility test, dynamic shear rheometer, bending beam rheometer, infrared red spectroscopy and fluorescence microscopy. Overall, the results indicated that the samples containing RAP and PET derived additives provided comparable performance compared to conventional binders and successfully demonstrated a novel approach to deal with two local waste recycling difficulties.

INTRODUCTION

Waste Plastic in Hong Kong

Being cost effective and easy to produce, plastic materials are extensively used around the world. The world production of plastics has experienced an exponential growth in recent times from 1.5 million tonnes in 1950 to 288 million tonnes in 2012 (ISWA, 2014). In the report published by the Hong Kong Environmental Protection Department (EPD) in 2016, 5.74 million tonnes of MSW in 2015 was generated in Hong Kong with 3.71 million tonnes disposed of at landfills and 2.03 million tonnes recovered through recycling. Waste plastics were the second largest waste category behind food waste with a total of 891 thousand tonnes generated in 2015. The rate of recovery of plastic waste was relatively uneven as 797 thousand tonnes (89.5%) of it was disposed at landfills and only 93.9 thousand tonnes (10.5%) were recovered through recycling. In terms of the sources of plastic materials disposed, carrier bags account for one-third while plastic bottles were the second biggest classifiable category. With regard to the type of polymer, Polyethylene (PE) and Polyethylene terephthalate (PET) were the major types of polymers found in the plastics waste which accounted for 16.8% and 4.6% of the disposed plastic respectively.

The trend of imbalance between plastics waste disposal and recovery has existed for several years. As indicated in Figures 1 and 2, the rate of disposal of plastics waste has shown a steady upward trend whereas the rate of recovery of plastics wastes indicates a significant decline from 2011 to 2015. The plans to reduce the amount of waste sent to landfills and increase the amount of waste being recycled has not been fully successful. Therefore, there is an urgent need to introduce new approaches in order to address this issue.

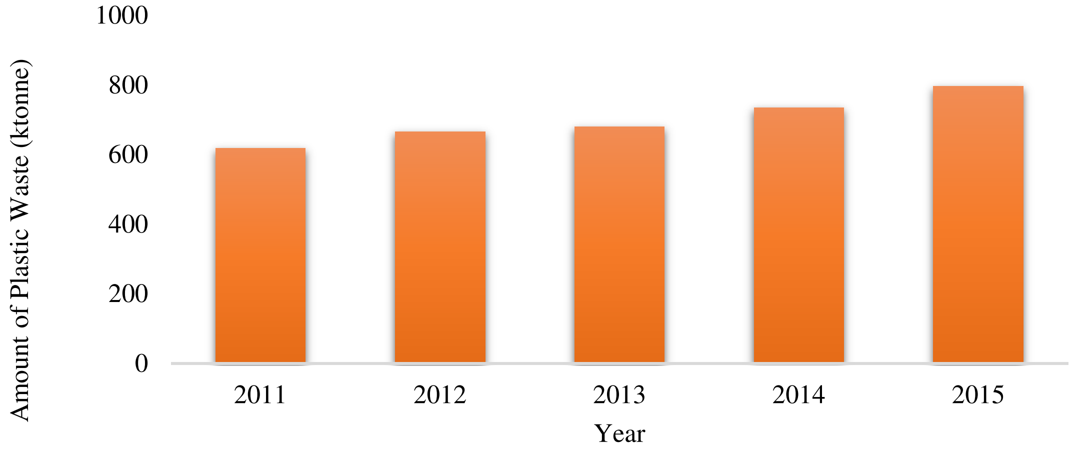


Figure 1. Plastic Waste Disposal Rate in Hong Kong from 2011 to 2015 (EPD, 2016)

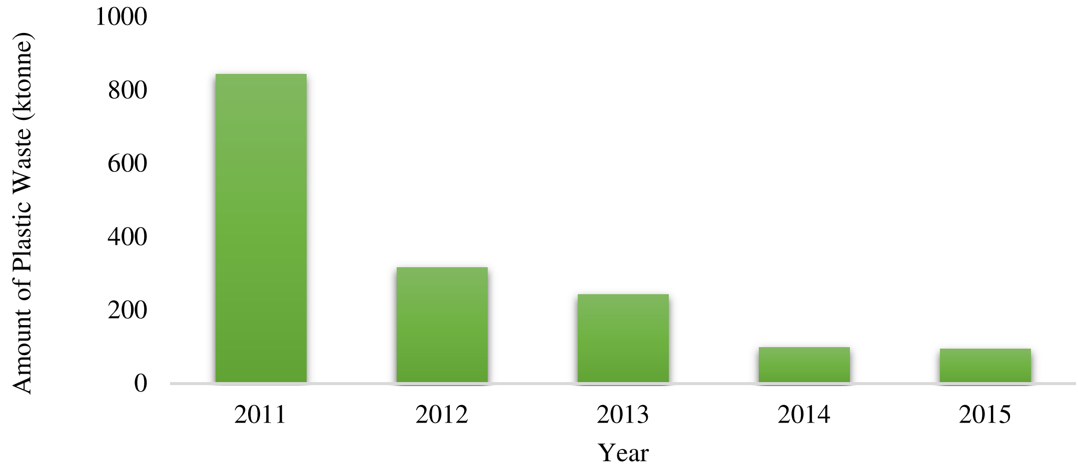


Figure 2. Plastic Waste Recovery Rate in Hong Kong from 2011 to 2015 (EPD, 2016)

Reclaimed Asphalt Pavement (RAP)

According to the EPD (2016), waste materials from road works are classified in the overall construction waste category. In 2015, 1.5 million tonnes of overall construction waste was generated and disposed at landfills. This amount of waste constitutes 28% of the total being disposed at landfills (Figure 3). One of the main constituents of the waste materials from road works is reclaimed asphalt pavement (RAP), which is the term given to removed or reprocessed pavement materials containing asphalt and aggregates. The reuse of these materials for road construction has been widely promoted due to its environmental and economic benefits both in Hong Kong and around the world. The recycling of RAP is also encouraged as it provides a holistic approach towards sustainable pavement systems. In the latest term contracts managed by the Highways Department in Hong Kong, 30% is the maximum allowable RAP content to be incorporated in bituminous mixtures. Around the world, there has been an observable increase in the usage of RAP in recent years and many highway agencies conducting their own research studies to increase RAP usage as well.

By nature, RAP is an old material and the binder in RAP is aged and stiffer, hence the incorporation of this aged binder to virgin HMA material results in a modified mix that is potentially also firmer. It has been well documented that the inclusion of high quantities of RAP notably increases the dynamic modulus of HMA mixes compared to those of virgin HMA mixes (McDaniel and Shah 2003, Pereira *et al.* 2004, Li *et al.* 2008). One of the main concerns prohibiting its widespread use is the lack of confidence in the long-term performance especially with regard to cracking and moisture damage (North Carolina Department of Transportation 2007, Materials Engineering and Research Office MTO

2008). The use of rejuvenators or additives, such as polymers, in RAP generally improves the deformation and stripping performance of HMA mixes (Kim *et al.* 2014), however when polymers are used in conjunction with RAP, the behaviour of the resulting mix can vary considerably depending on the type of polymer used.

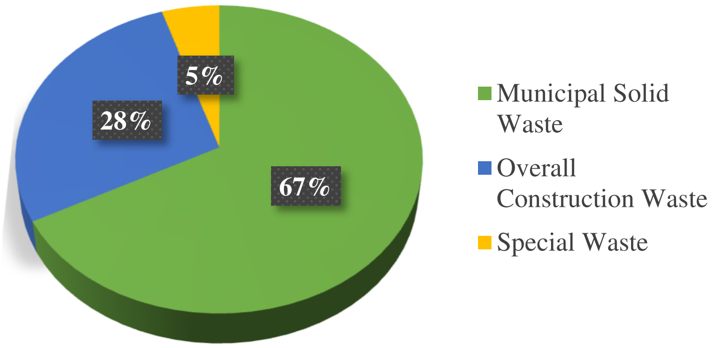


Figure 3. Composition of Overall Construction Waste Disposed at Landfills in 2015 (EPD, 2016)

Concept- Combining Waste Plastic and RAP as Sustainable Pavement Materials

Meeting waste management challenges in the 21st century continually requires inventive and decisive approaches in terms of engineering and policy. Waste plastic and RAP are two waste management challenges in Hong Kong that need to be urgently met. The objective of this study is to evaluate the feasibility of using waste plastic additives in high RAP bituminous mixes. Polyethylene Terephthalate (PET), a thermoplastic polyester which constitutes 18% of the total polymer produced worldwide (Ji 2013) and commonly used to make plastic bottles in Hong Kong, is used as the waste plastic additive in this research. The usage of waste PET as an additive for asphalt road material has been studied previously although it can be regarded to be at an early stage. In prior studies, PET waste was generally added to the asphalt mixture in a dry process, i.e. used as aggregate in the asphalt mixture in order to improve the resistance to permanent deformation, Marshall stability, stiffness and fatigue life of road pavement (Ahmadinia *et al.* 2011, Moghaddam *et al.* 2014). Apart from this, asphalt was modified with a number of additives derived from PET waste by aminolysis and glycolysis reaction from which it was found to improve the fatigue properties, Marshall stability and moisture resistance depending on the asphalt and additive contents (Padhan *et al.* 2013, Gürüa *et al.* 2014). These initial studies have indicated that the PET additives have significant potential to improve the stripping characteristics and overall performances of asphalt mixtures. It is likely that PET additives act similar to commercial rejuvenating agents and hence would be suitable to be incorporated into high RAP mixes to improve moisture damage and cracking properties. A sustainable pavement system using both waste plastics and RAP would create an unprecedented recycling outlet for these waste materials and help alleviate the pressure of disposal.

Figure 4. Conceptual Diagram for Using Waste PET and RAP in Road Pavement Design

MATERIAL PREPARATION AND RESEARCH METHODOLOGY

Materials

Bitumen with a penetration grade of 60/70 was used as the virgin binder to produce the PET modified binder samples. The waste PET bottles were collected locally, all PET bottles were first cleaned and dried, and then cut into small pieces of approximately 5 mm by 5 mm. The chemicals used, triethylene tetramine (TETA) and xylene, were of laboratory reagent (LR) grade. The RAP was obtained from the Highways Department, Hong Kong. It was obtained from a 10 mm wearing course cold milling and was reported to be of low to moderate ageing level. The RAP binder was extracted using the solvent extraction method as per AASHTO T164, 2014.

Synthesis of PET Additive

A three-necked 500 ml round bottomed flask equipped with a heating mantle, overhead stirrer, water condenser, nitrogen gas sparging tube and a thermos well pocket containing a thermometer was charged with 30 g of PET, 100 g xylene, 60 g of triethylene tetramine (TETA) in the presence of nitrogen gas. The mixture was heated at around 130 ºC to 140 ºC to for eight hours under reflux. The solution turned homogeneous as the PET degradation completed. The PET degradation was confirmed by IR analysis which showed the disappearance of ester group peak at 1735 cm^{-1} and the formation of amide peaks at 1637.6 cm^{-1} and 1544.4 cm^{-1} for TETA amide (Figure 5). The surface structure of PET additives under the scanning electron microscope (SEM) is also shown in Figure 6. At the end of the reaction the polyamines and glycols are recovered under vacuum. The resulting product is a residue recovered in quantitative yields at ambient temperatures (Figures 7&8).

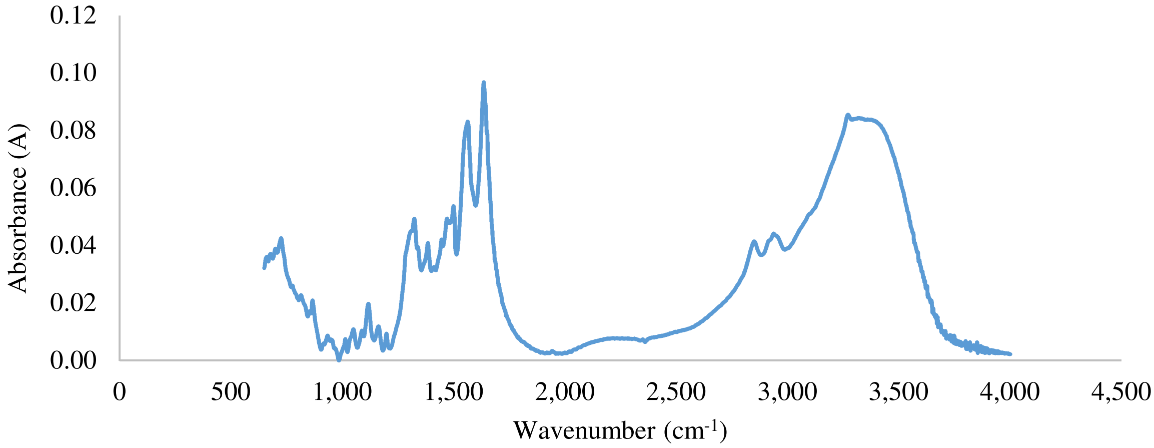


Figure 5. FTIR Spectrum of PET Additives

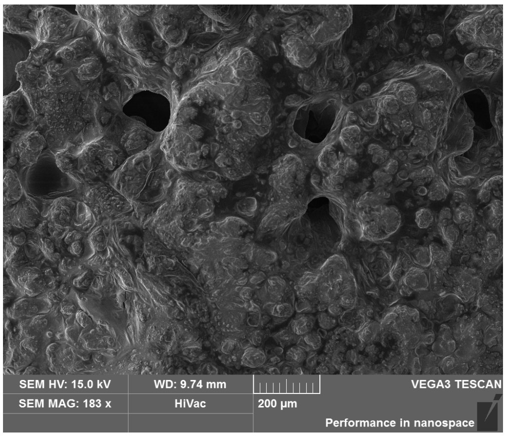


Figure 6. SEM Image of PET Additive

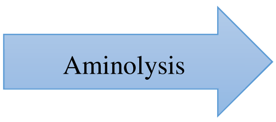


Figure 7. Scrap PET

Figure 8. PET Derived Additive

Preparation of Modified RAP and PET Asphalt Binder

The optimum percentage of PET additives used for this study was chosen based on prior work carried out by Padhan *et.al* (2013) and extensive antistripping tests. Two control samples of virgin binder, virgin binder with 2% PET additive (PET modified binder) and PET modified binder mixed with 15%, 25% and 40% RAP binders were prepared using a high shear mixer at around 150 °C for 2 hours at a mixing rate of 4000 r/min. Table 1 shows a sample criterion for the selection of the optimum percentage of PET additive used for this research study. The percentage (%) of antistripping characteristics in Table 1 refers to the amount of binder that has not been stripped from the local granite based aggregate after hot water immersion tests in accordance to ASTM standards (ASTM-D3625).

Table 1. Selection Criteria for % of RAP binder based on Hot Water Stripping Tests

Binder Composition	Percentage (%) of Antistripping
100% RAP binder	40 to 45
25% RAP binder + PET (1%) modified binder	65 to 70
25% RAP binder + PET (1.5%) modified binder	75 to 80
25% RAP binder + PET (2%) modified binder	85 to 90
15% RAP binder + PET (2%) modified binder	90 to 95
40% RAP binder + PET (2%) modified binder	75 to 80

RESULTS AND DISCUSSION

The flowchart below shows the experimental program of this study. To better understand the working mechanism of the PET additive in the asphalt binder, chemical characterisation tests such as Fourier Transform infra-red (FTIR) and Fluorescence microscopy (FM) tests were also conducted in addition to conventional binder tests.

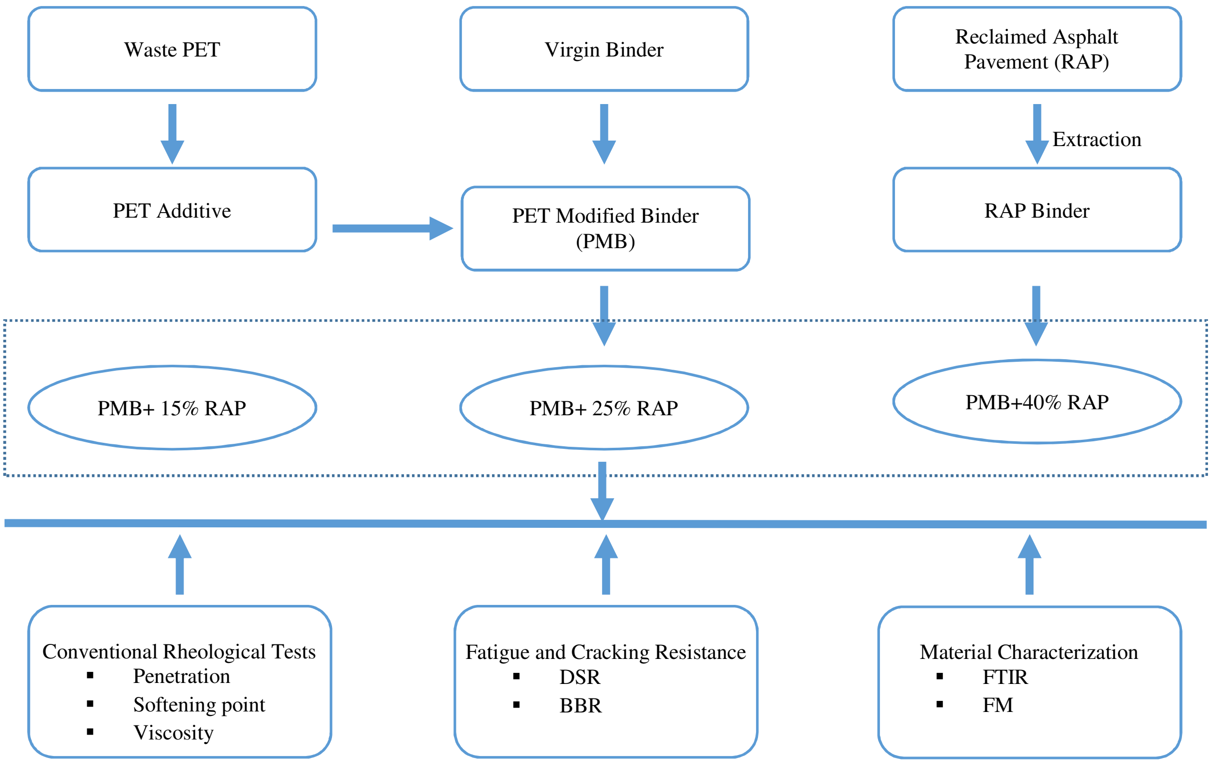


Figure 9. Flow Chart of the Experimental Program

Table 2 illustrates the conventional binder properties of the various samples. The penetration test at 25ºC, the softening point tests and the ductility tests were conducted for the RAP binder, virgin binder and the PET modified binders.

Table 2. Conventional Binder Properties

Properties	Virgin Binder	RAP Binder	PET Modified Binder	PET Modified Binder + 15% RAP	PET Modified Binder + 25% RAP	PET Modified Binder + 40% RAP
Penetration (0.1mm, 25ºC,100g, 5s)	69	40	54	61	57	52
Softening Point (ºC)	48	57	49	50.5	51	52.5
Ductility (cm)	100+	85	100+	100+	100+	100+

Viscosity

The viscosity test was carried out using a rotational viscometer at various temperatures to ascertain the change in viscosity of the binders. As predicted, the aged binder has a much higher viscosity than the virgin material. This was expected as the asphalt binder in RAP loses light components during service. The PET modified binders with RAP have comparable viscosity to the virgin binder at mixing temperature (150ºC).

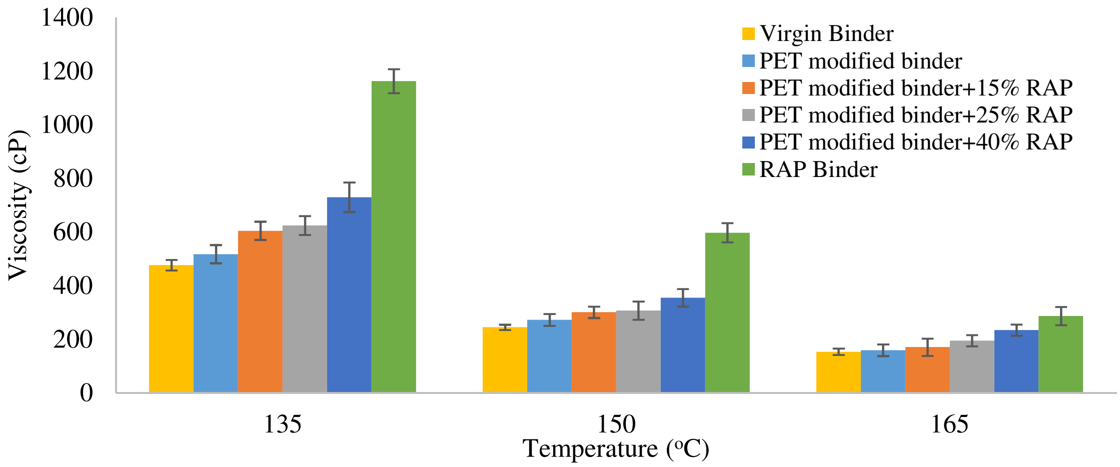
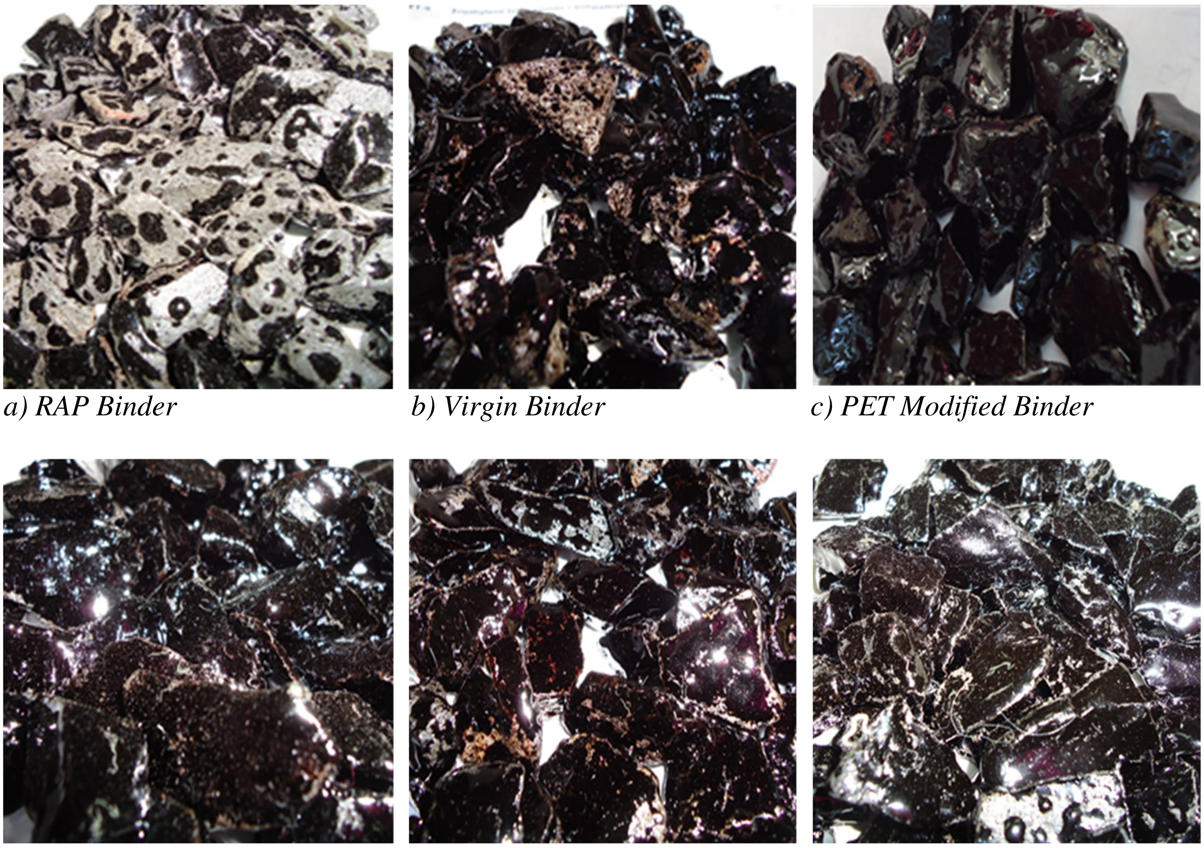


Figure 10. Viscosity of Various Asphalt Binders

Antistripping Properties

The most commonly used tests to ascertain the antistripping properties include the hot water immersion test and the Marshall stability test. The samples were tested for the hot water immersion tests (ASTM D3625) and some of the results are shown in the Figure 11. The PET modified virgin binder is the most resistant to stripping owing to the addition of the antistripping additives. The addition of 15% and 25% RAP binder did not deteriorate the stripping characteristic of the PET modified binder to a significant extent and was even comparatively better than the virgin asphalt. Thus it can be inferred that the addition of the PET additives significantly improves the antistripping characteristics of samples containing RAP binder.



a) RAP Binder *b) Virgin Binder* *c) PET Modified Binder*

d) PET Modified Binder with 15% RAP *e) PET Modified Binder with 25% RAP* *f) PET Modified Binder with 40 % RAP*

Figure 11. Photos Taken after Hot Water Immersion Testing

Dynamic Shear Rheometer (DSR) Test

The DSR test was conducted to evaluate the permanent deformation and fatigue cracking properties of the different mixtures. Physical property measurements were made for the original binder samples, rolling thin film oven (RTFO-aged) samples and the pressurized ageing vessel samples (PAV). In all DSR tests, the PET modified binders showed higher $G^*/\sin\delta$ (rutting factor) values as compared to the virgin binder (Figures 12&13). When RAP binder was added to the PET modified binder, there is a noticeable increase in $G^*/\sin\delta$ at all temperatures, i.e. 58 °C, 64 °C ,70 ºC, 76 ºC. This indicates that RAP modified PET binder has higher service temperature than PET modified binder and virgin binder. When considering the DSR results for the PAV aged samples (Figure 14), the value of $G^*\sin\delta$ is lower for the PET binder modified with RAP indicating better performance to fatigue cracking as compared to the virgin binder.

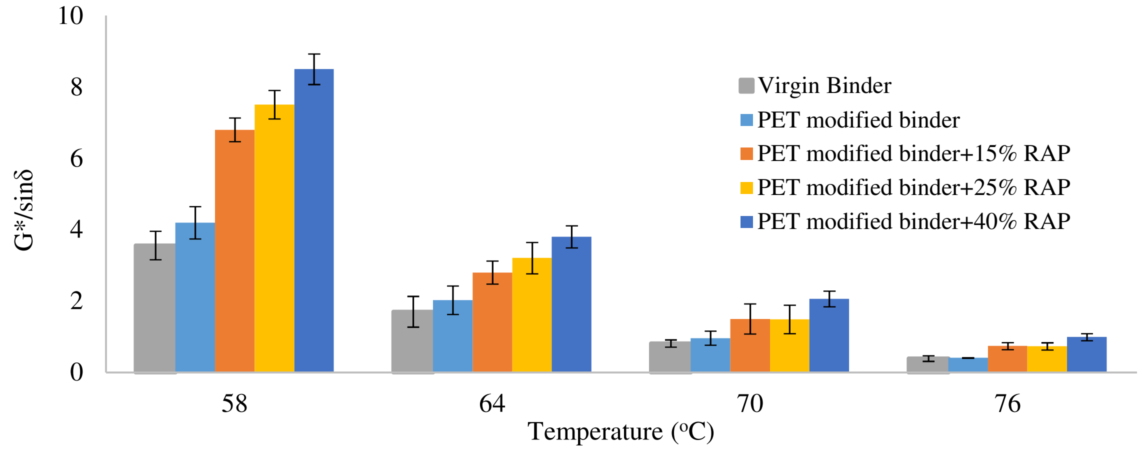


Figure 12. G/sin*δ vs Temperature for Original Binder *Samples*

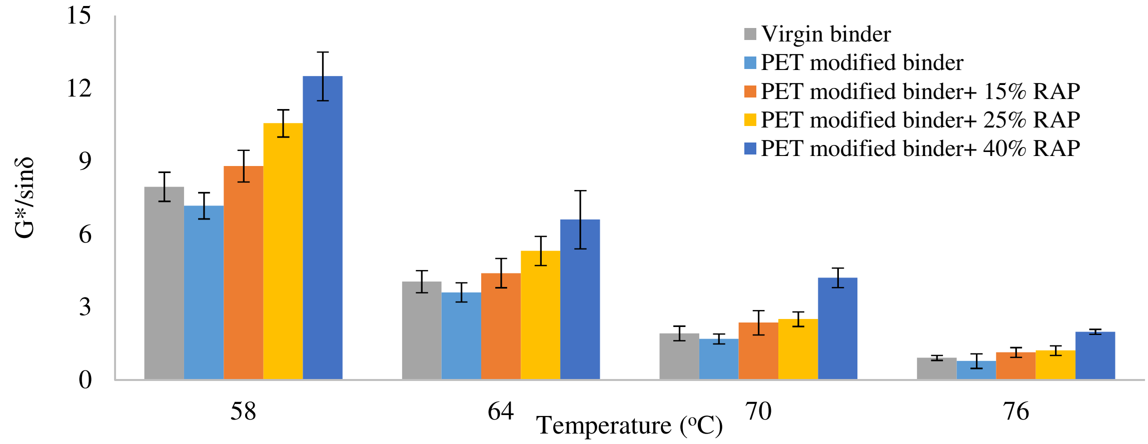


Figure 13. G/sin*δ vs Temperature for *RTFO-aged Samples*

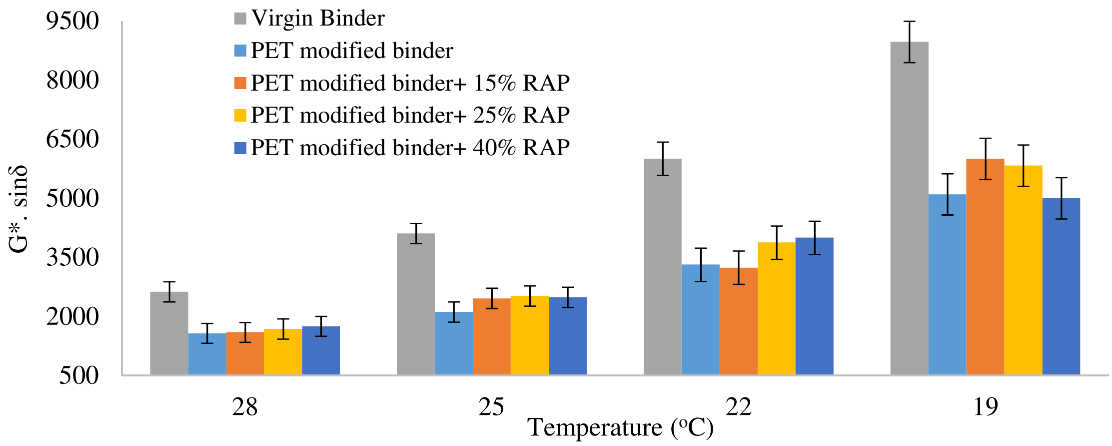


*Figure 14: G*sin*δ vs Temperature for *PAV samples*

Bending Beam Rheometer (BBR) Test

The BBR test was carried out at the required low temperatures of -6 ºC to -18 ºC for testing the low temperature cracking properties that is generally a matter of concern for binders containing RAP components. Two parameters were obtained through this test; stiffness and the rate of change of stiffness with time (m-value). It can be noted that all the binder samples performed similarly and met the creep stiffness specifications up to -12 °C, hence meeting the low temperature specifications for - 22 °C in terms of creep stiffness (Figure 16). Similar trends are also observed for the m-values. A high m-value (> 0.3) is generally desired, as the temperature changes and thermal stresses accumulate, the stiffness will change quickly. A quick change in stiffness will cause the binder to shed stresses which can build up to a level where low temperature cracking could occur. It can be inferred from the results that the addition of the RAP binder does not severely deteriorate the performance of the PET modified binders (Figures 15&16).

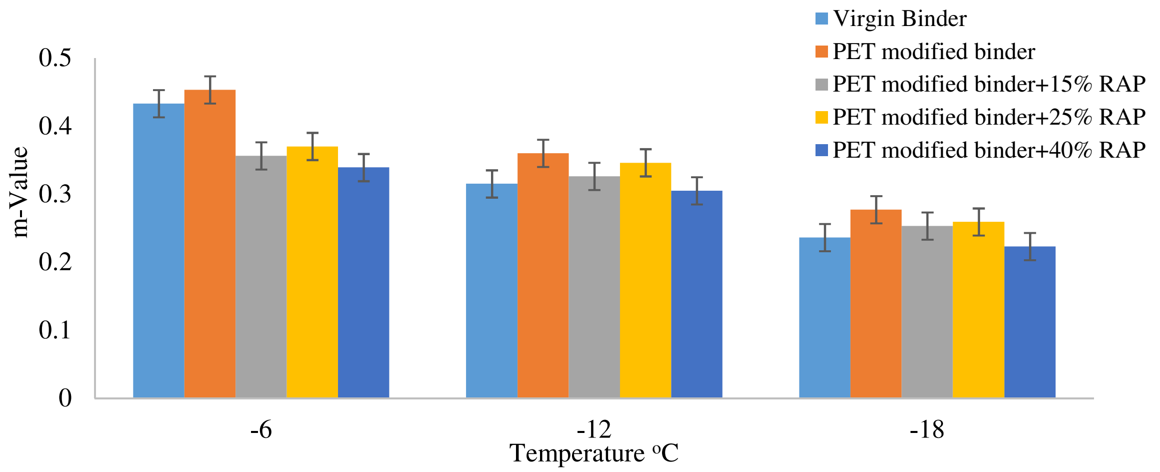


Figure 15. m-value Results vs Temperature

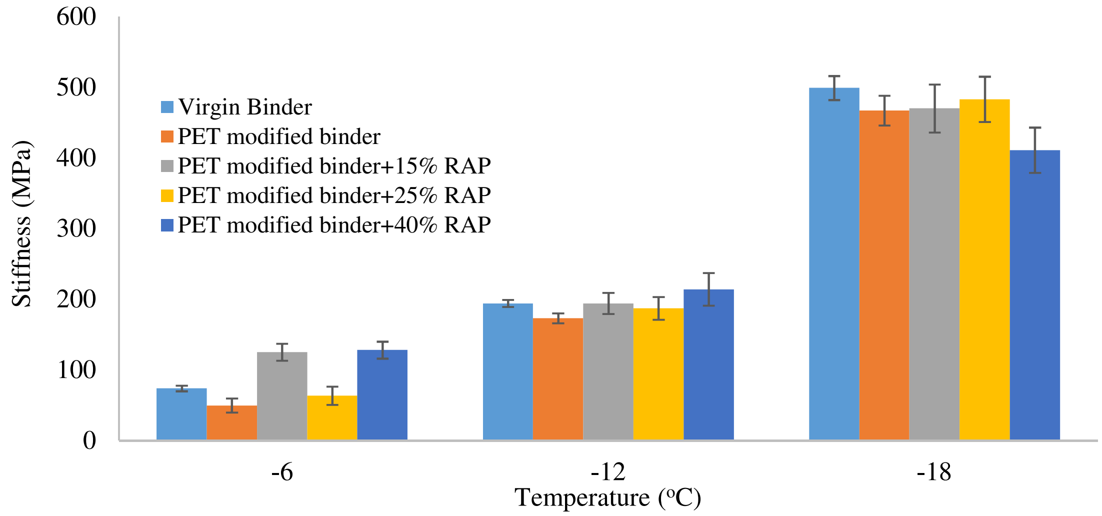


Figure 16. BBR Test for Stiffness

Fourier Transform Infrared Spectroscopy (FTIR) Analysis

The FTIR analysis was conducted on the samples to characterize the various functional groups in the original and modified binder samples. The aged RAP binder displayed physical and chemical changes as it was subjected to a thermal oxidative process over time (Figure 17). This can be attributed by the loss of volatiles or specimens of low molecular weight, or even the formation of hydrogen bonds. The formation of sulphoxide groups characterized by the band at 1030 cm^{-1} frequency (S=O stretching) was also noted. The absorption at the frequency 1160 cm^{-1} can be attributed to the anhydride groups formed after oxidation. A small number of carbonyl groups were also observed at a frequency of 1700 cm^{-1} in the case of the RAP Binder. However, the C=O peak is diminished when RAP binder is blended with PET modified binder. It can also be inferred from the FTIR tests that the amount of oxidative products has been reduced in the PET modified binder with RAP indicating that the combined RAP+PET binder behaves similar to unaged binder.

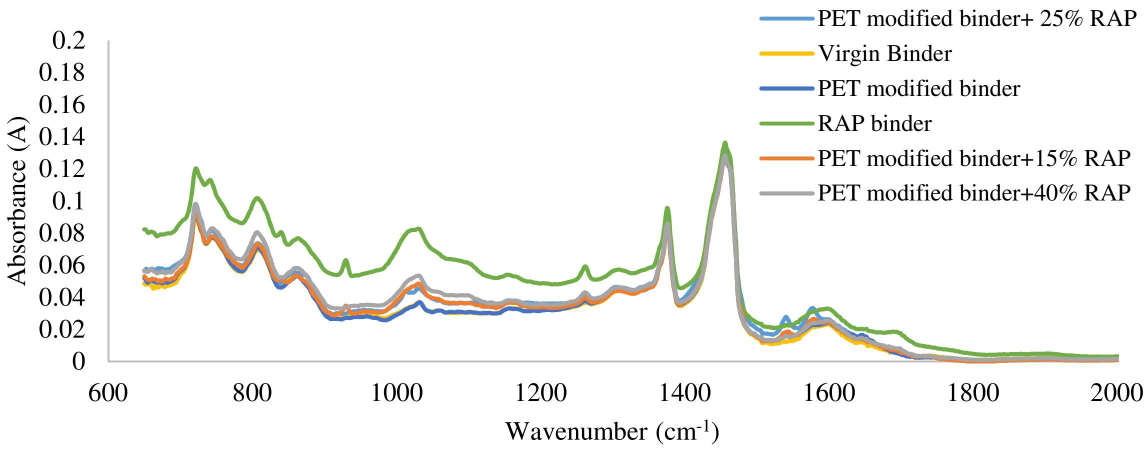


Figure 17. FTIR Spectroscopy Analysis of Mixes

Fluorescence Microscopy (FM)

In addition to changing the rheological and chemical nature of RAP binder, PET additives also effect the morphology (structure) of the asphalt binder. Fluorescence microscope (FM) is a microscopic method, which enables the observation of polymer and additives distribution in asphalt. It can be seen from the FM image (Figure 18) that mixture of PET modified binder is homogenous and the dispersed PET is represented by the dotted particles. The mechanism is that the aromatic phase of asphalt is usually the most fluorescent phase, and once polymeric additives absorb aromatics, they become bright when exposed to fluorescence (Polacco *et al.* 2015). It can be observed from the FM images that the PET additive has uniformly dispersed inside the asphalt mixture. The different morphologies seen in Figure 18 are a function of the swelling potential of the PET additives, the nature of the virgin asphalt (composition of the maltenes fraction) and the asphalt-PET additives compatibility. The different chemical and morphological nature of the PET additive inevitably effects the rheological characteristics of these modified binders.

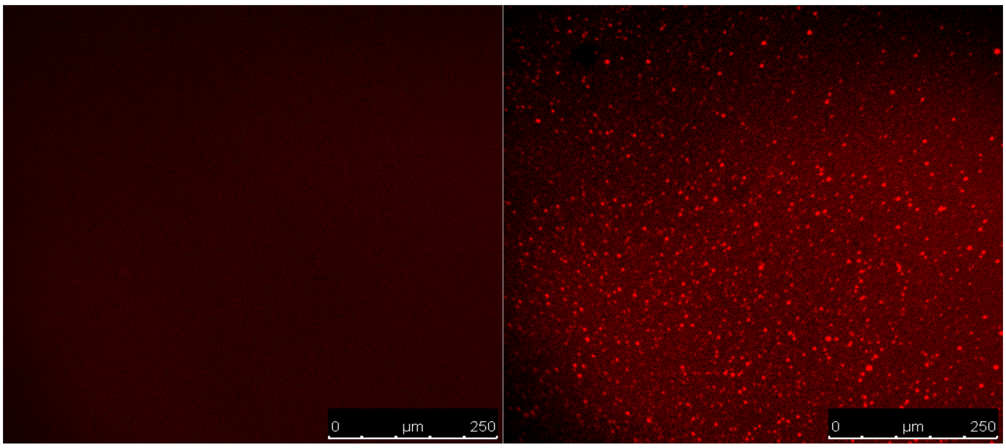


Figure 18. Fluorescence Microscope Comparison of Virgin Binder and PET Modified Binder

FINDINGS AND CONCLUSION

The aim of this study was to develop a novel method to recycle waste PET bottles and incorporate them into pavement mixtures containing reclaimed asphalt pavement (RAP). The PET additive used was shown to be suitable to be incorporated in aged binders containing RAP, providing better or equivalent performance to conventional virgin binder. The salient features of the laboratory tests are listed below:

- Based on the stripping tests, the PET additive would be suitable to be incorporated into high RAP mixes as it considerably reduces stripping characteristics of RAP binder and shows better antistripping characteristics than virgin binder.
- The DSR tests indicated that the samples containing RAP binder performed considerably better, improving the fatigue performance at medium temperatures and rutting characteristics at high temperatures.
- The addition of the PET additive also enhanced the low temperature performance of the modified binders with RAP which was then comparable to virgin binder.
- From the FTIR spectroscopy studies, it was observed that the amount of oxidative products were reduced in the modified binder with RAP indicating the combined mixture behaves similar to unaged binder or that the PET additive could have rejuvenating properties.
- It was established from the fluorescence microscopy images that the mixture of PET modified binder is homogenous which in turn confirms the observed rheological properties of the samples.
- Future research will be focused on conducting mixture studies to ascertain the viability for field tests.

Overall, this study has successfully demonstrated an innovative approach to deal with two waste difficulties: waste plastic and RAP, and provides a competitive technology to meet this locally relevant recycling challenge.

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