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Evaluating the Rheological Properties and ageing Resistance of Waste PET-Modified Bitumen Binder

Nuha Mashaan*, Amin Chegenizadeh & Hamid Nikraz

School of Civil and Mechanical Engineering, Faculty of Science and Engineering, Curtin University, Bentley WA 6102, Australia.

*Corresponding author: nuhas.mashaan1@curtin.edu.au

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ABSTRACT

Reusing waste plastic can significantly contribute to saving the environment from further pollution and the economy from extra expenses. Waste plastics have a vital role in the current asphalt applications. Trial sections of roads made with waste plastic modified asphalt during 2019 in Australia, highlighted the significant importance of laboratory assessments; however, they are yet to be conducted. This study investigates the impact of using local waste Polyethylene Terephthalate (PET) plastic in bitumen binder class C320, which is a common bitumen type used for local road surfacing in Australia. The evaluation of numerous contents of waste PET modified bitumen is carried out before and after ageing conditions. Dynamic Shear Rheometer (DSR), Rolling Thin Film Oven (RTFOT) and Pressure ageing Vessel (PAV) tests were utilised to examine the rutting, fatigue, and ageing of bitumen binder. The results displayed that the use of waste PET display improved performance in terms of rising the complex shear modulus and reducing the bitumen's susceptibility to deformation at high temperatures. Furthermore, 8% of waste PET increases the fatigue cracking resistance as it shows a low fatigue factor. Further rutting and fatigue tests on asphalt mixtures samples are recommended to better understand the mechanical properties of waste PET plastic on modified C320 asphalt mixtures.

Keywords: waste plastic; rheology; rutting; fatigue; modified bitumen; environmental impact.

INTRODUCTION

With the developing world's growing population, the quantity of waste generation proliferates. This amount of waste causes a considerable rise in the fee of waste dumping and fills future sites for land fields. Presently, Australia has faced the essential concern of recycling waste plastic. On 1 January 2018, China enforced a prohibition on the importation of waste plastic (O'Farrell 2018). The ban caused numerous corporations to abandon waste collection agreements and start stockpiling waste as there is nowhere to deposit this waste safely. This issue seems, to a great extent, placed Australia's recycling industry in a crisis. Therefore, local waste of plastic needs to be dumped in stockpiles which drastically increases the level of

hazards and pollution in Australia. As a result, local councils will have to find strategic ways of recycling the accumulated waste that will become a more significant issue in the coming years (O'Farrell 2018).

The application of waste materials instead of new materials in the construction of roads has two significant benefits. One is the substantial savings and reduced costs, and the second is the cutting down of waste that will be disposed in the landfills. Thus, the future development of using waste plastic in bitumen modification will consider the enhancing properties of the mixture (Dalhat & Al-Abdul Wahhab 2017; Mashaan et al. 2019a, 2019b; Piromanski et al. 2020; Zakaria 2020; White 2020; Hamedi et al. 2020).

Modification of the bituminous mix with waste plastics seems to have great potential for the efficient use in flexible pavements design to enhance their active service or minimise the layer thickness of its wearing course or base layer (Ameri et al. 2017). The application of waste plasticmodified bitumen enhances the pavement's service life length, especially in severe conditions such as parking areas tolerating heavy traffic loads, deformed road bases, and stress-relieving interlayers. It has been proven to meet the requirements for optimal performance in modern pavement construction and coatings; also, it appears to be a practical, logical, and economical approach compared to other approaches. Polymer modified bituminous mixtures have an extensive choice of applications in most countries (Ameri et al. 2017; Costa et al. 2019). Adding the waste plastic polymer to the bituminous mixture increases its stiffness and improves its non-susceptibility to temperature fluctuations in different regions and climates. This feature, in turn, raises the level of resistance of the mixture to rutting (Costa et al. 2019; H1N1SL1OĞLU & AĞAR 2004). Recycling waste plastics can significantly contribute to saving the environment from further pollution, and the economy from extra expenses. Generally, adding plastic polymer into asphalt results in an enhancement in the fatigue and rutting resistance of bitumen binder, due to the improvement in bitumen's temperature susceptibility and stiffness.

Currently, in Australia, it is costly to use asphalt mixtures modified by commercial polymers for road pavement purposes. Therefore, one significant way of reducing costs would be to use waste plastic polymer as a modifier. To the authors' knowledge, there is no substantial laboratory assessment on the effects of local waste plastic modified Australian wearing course materials on the engineering properties of bitumen binder. Consequently, the current research study aims to investigate the use of recycled plastic modifiers for asphalt production by using Australian materials and test methods. Along with C320 Australian asphalt used as the control for relative performance, this study endeavours to examine the effects of local waste PET plastic to enhance the bitumen binder rutting and fatigue properties. The study also aims to discuss the previous studies and background issue of using waste plastic as an asphalt-modifier in pavement engineering, as illustrated in the next section.

BACKGROUND ON USING WASTE PLASTIC POLYMER IN PAVEMENT ENGINEERING

With the developing world's growing population, the quantity of waste generation proliferates. This amount of waste causes a considerable rise in the fee of waste dumping and fills future sites for landfills. Substantial work has been conducted into reusing waste through re-usable by-products (Sara et al. 2017). Reusing is a type of recycling which reduces the amount of waste, cost for transportation and energy, and lessens the demands for new resources (Tam & Tam 2006). Studies are currently researching the advantages of reusing waste plastic material in an economically and environmentally ecological way (Huang et al. 2007; Aubert et al. 2007; Abo El-Naga 2019; Esfandabad et al. 2020; Mashaan et al. 2021). There have been many investigations into the consequences of reusing hazardous waste on the construction of pavement material's performance, strength, and environmental impact (Xue et al. 2009; Mashaan & Karim 2013). Road pavements start experiencing functional deterioration once they undergo heavy traffic or the freezing of groundwater during the cold season. Deterioration can include fatigue or alligator cracks, edge cracking, grade depressions, slippage and block cracking, potholing, ravelling, shoving, stripping, and rutting.

According to investigations' reports (Asare et al. 2019; Santos et al. 2020; Jamshidi & White 2020; Hake et al. 2020), the use of waste plastic as a modifier in asphalt construction would be an effective way of protecting the environment from additional contamination and helping the economic system, by sparing additional cost. Moreover, utilizing plastic polymer into asphalt improves the bitumen's temperature susceptibility and stiffness. This enhancement of bitumen re-sults in an enhancement in the rutting and fatigue cracking resistance of asphalt pave¬ment (Haider et al. 2020; Al-Haydari & Al-Haidari 2020; Kumar & Khan 2020).

Modifying and advancing the properties of bitumen and asphalt mix by using certain additives, such as plastic polymers, is one way of boosting the service life of road surfaces (Casey et al. 2008; Al-Hadidy & Yi-Qiu 2009a, 2009b; Ozen et al. 2008). Studies that have been conducted to classify bitumen modifiers according to their composition have been categorised into several groups. These include polymers elastomeric, polymers plastomeric, fibre, and crumb rubber, as illustrated in Table 1. These additives vary considerably in their physical and chemical properties, which have a wide variety of influences on the performance of asphalt concrete. Additives render the mixture stiff, especially in hot conditions, and less stiff at colder temperatures. These materials control the elasticity of the mixture in normal temperature conditions.

The large and uncontrolled amount of PET plastic bottles produced in recent decades have contributed to the creation of serious environmental problems, mostly because of hygienic consideration, in that these plastics are not reusable for refilling. Hence, nearly all the produced bottles are disposed of as waste plastic materials whose decomposition and return to nature is outside of the lifetime of the current generation (Mahdi et al. 2007). Recycling these PET bottles can be an effective and immediate solution to this problem in industrial and consumer societies.

During the last decade, there were only a few studies on using a plastic polymer as additive and asphalt modifier as illustrated in Tables 2 and 3. These studies show significant and encourageing results of using plastic in asphalt, nevertheless; there is lack of studies to show the substantial contribution of using high contents of more wasted plastics like PET and High-Density Polyethylene (HDPE) in asphalt modification. Al-Hadidy (2009a,2009b) had attempted to study a kind of Polyethene that was used as an additive in asphalt mixtures. The studies that were specifically concentrating on the modification of asphalt mixtures through the application of Polyethene are inadequate (Al-Hadidy & Yi-qiu 2009a, 2009b). The studies covering Polyethylene reinforced asphalt mixtures and binders form only a small portion of the current publications. There is still a necessity for further studies focusing specifically on this topic.

A joint research by Awwad & Shbeeb (2007) investigated the results of polyethylene polymers' engagement to improve the properties of asphalt mixtures. Their study was conducted to determine the best polyethylene type and the proportion to be used in the asphalt mixture to obtain the optimal result. Hence, authors applied two types of polyethylene to the aggregate coating, specifically, High-Density Polyethylene (HDPE) and Low-Density Polyethylene (LDPE), respectively. The addition of the plastic polymers to the mixture was carried out in two forms: ground and unground. The produced mixture samples displayed that the ground HDPE imparts better engineering properties to the resulting mixture. The most appropriate percentage of the modifier suggested by researchers is 12% by weight of bitumen. The results of this experiment further confirm that the introduced HDPE can contribute to the enhancement of mixture stability, a slight increase of the air voids in the mineral aggregate in it, and the reduction of the asphalt mixture density (Awwad & Shbeeb 2007). Several studies revealed that using recycled waste plastic materials primarily composed of LDPE in bituminous mixtures resulted in a significant enhancement of its stability. Approximately 2.5 times greater than the stability of the unmodified-mixtures while decreasing its density (Zoorob & Suparma 2000; Ho et al. 2006; Costa et al. 2019).

Hinishoğlu & Ağar (2004) used other kinds of waste plastic materials with HDPE to modify binders with various blending temperatures, time period, and HDPE percentage. For this experiment, they used Marshall Stability, Marshall Quotient, and Marshall Flow. Investigations concluded that 4% of HDPE at 165 °C mixing temperature blended continuously for 30 minutes is the best condition for meeting all requirements. As a result, the new condition applied to this experiment, the percentage of the Marshall quotient was raised by 50% as compared to the control mixture. Furthermore, the researchers noted that resistance of the HDPE modified bituminous mix against serious deterioration and deformation was significantly increased.

Additionally, Polyethylene utilization of the porous asphalt might result in significant improvements in its oilresistance properties. During the experiments, it had noted that adding plastic-polymers would significantly enhance the cracking and failure resistance of the modified asphalt mixes (Tasdemir et al. 2009; Attaelmanan et al. 2011). Moreover, it was also revealed that Polyethylene could contribute to substantial enhancement in terms of bending fatigue destruction-resistance and anti-stripping properties of the dense-grade asphalt mixtures.

TABLE 1. Classifications of asphalt additives

Type of additive	Authors & studies	Findings						
Crumb rubber/ waste tyre rubber	Moreno et al. (2011); Aflaki & Memarzadeh (2011); Hamid (2010); Mashaan & Karim (2013); Ali et al, (2013); Mashaan et al. (2014); Shen & Amirkhanian (2005); Bahia et al. (1994).	 Improved rheological properties. Improve ageing resistance. Improve stability Develop a bond between aggregate and binder. Improve fatigue life. Better rutting resistance. 						
Elastomers: -Styrene- butadiene-styrene (SBS).	Al-Hadidy (2018); Ozen et al. (2008); Khattak & Baladi (2001); Ahmed (2007); Widyatmoko & Elliott (2008)	 -Increase stiffness - Resist fatigue cracking. - Resist thermal cracking. 						
Fibre: - Polyester - Fibre glass	Wu et al. (2008); Putman et al. (2004); Huang (1996).	Improves tensile strength - Improve cohesion - Improve fatigue life. - Improve durability - Improve stability of mix.						

		INDEL 2. Types of p	lastic and properties		
References	Type of plastic waste	Shape	Size (mm)	Melting point °C	Specific gravity / density (g/cm3)
Zoorbo & Suparma (2000)	LDPE	Pellet	5.00-2.36	140 °C	0.92
Hinislioglu & Agar (2004)	HDPE	Powder	2 mm		0.935 g/cm3
Awwad & shbeeb (2007)	HDPE & LDPE	Grinded & Not grinded	2-3 mm	HDPE=125°C LDPE=110°C	HDPE=0.035 LDPE=0.033
Al-Hadidy & Yi- qui (2009a)	PP (virgin)	Powder	Not given	Not given	0.82gm/cm3
Ho et al. (2006)	PE LDPE wastes	PE: wax LDPE: pellet and shredded	Not given	Not given	Not given
Al-Hadidy & Yi- qui (2009b)	LDPE (virgin)	Grinding to powder	Not given	113.2C	0.9205gm/cm3
Casy et al. (2008)	Wastes PP HDPE LDPE	Mulch & powder \powder	Not given	LDPE: 110C HDPE: 131C	Not given
Vansudevan et al. (2012)	PE PP PS	Foam, powder	Not given	Not given	Not given
Attaelmanan et al. (2011)	HDPE (virgin)	Pellet	Not given	149C	0.9430gm/cc (density)
Modarres & Hamedi (2014)	PET	PET chips were crushed and sieved	0.425–1.18	Not given	Not given
Khan et al. (2016)	LDPE / HDPE + CR	Powder	0.15-0.75 mm	Not given	Density LDPE: 922kg/m3 HDPE: 961kg/m3
	TABLE 3.	Properties of asphalt mix	using plastic and n	najor findings	
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Hinislioglu & Agar (2004)	HDPE	Powder	2 mm		0.935 g/cm3
Awwad & Shbeeb (2007)	HDPE & LDPE	Grinded & Not grinded	2-3 mm	HDPE=125°C LDPE=110°C	HDPE=0.035 LDPE=0.033
Al-Hadidy & Yi- qui (2009a)	PP (virgin)	Powder	Not given	Not given	0.82gm/cm3
Ho et al. (2006)	PE LDPE wastes	PE: wax LDPE: pellet and shredded	Not given	Not given	Not given
Al-Hadidy & Yi- qui (2009b)	LDPE (virgin)	Grinding to powder	Not given	113.2C	0.9205gm/cm3
Casy et al. (2008)	Wastes PP HDPE LDPE	Mulch & powder \powder	Not given	LDPE: 110C HDPE: 131C	Not given
Vansudevan et al. (2012)	PE PP PS	Foam, powder	Not given	Not given	Not given
Attaelmanan et al. (2011)	HDPE (virgin)	Pellet	Not given	149C	0.9430gm/cc (density)

TABLE 2. Types of plastic and properties

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Modarres & Hamedi (2014)	PET	PET chips were crushed and sieved	0.425–1.18	Not given	Not given
Khan et al. (2016)	LDPE / HDPE+ CR	Powder	0.15-0.75 mm	Not given	DensityL DPE: 922kg/m3 HDPE: 961kg/m3

METHODOLOGY

SAMPLE PREPARATION

The bitumen binder used in the current study is C320, which is supplied by Sami bitumen company WA, Western Australia. The physical properties of C320 in terms of viscosity at 135 °C, penetration at 25 °C, viscosity after Rolling Thin Film Oven test (RTFOT) at 60 °C are 0.5 Pa.s, 40 dmm, 260 Pa.s, respectively. Waste plastic bottle (PET) was collected, washed, and ground to a size less than 0.425 mm to be used as a modifier. Figure 1 shows the materials used in this research study.

The plastic-modified binder samples were prepared using the high shear mixer to mix the C320 with various content of plastic PET of 0%, 4%, 6%, and 8% by weight of bitumen. Based on the data of several trial mixtures, the choice of mixing conditions of temperature, time, and mixing velocity was 180°C, 40 minutes and 2000 rpm, respectively. The method of setting up the plastic polymermodified bitumen began at the low shear blend of 700 rpm for first 10 minutes to ensure less air void will be in the blend; after that point, the speed of blender is raised to 2000 rpm for 30 minutes. Tests of modified and unmodified binders were tested utilizing the DSR to determine the viscoelastic behaviour and to investigate the rutting and fatigue resistance when ageing. However, to assess the temperature susceptibility of the waste plastic modified binder, a broad scope of temperatures was picked and recorded as 50 °C,58 °C,60 °C,64 °C, and 70 °C. To simulate the ageing during construction, the RTFO method was used. The test was carried out to replicate the ageing process when the bitumen leaves the depot and is laid onsite. The RTFO test followed A/NZS 2341.10:2015. Both aged and unaged binders will provide insight into the ageing effects that waste PET polymer has on bitumen by using the ageing index (Ali et al. 2013). To imitate the ageing during service and long-term maintenance, the pressure ageing vessel was set to the conditioned temperature 100 °C and air pressure of 2.1MPa for 20 hours (AASHTO R28:2009).



FIGURE 1. Shows the materials used in this research study of waste PET plastic (A), plastic after grinding(B) and binder mixer (C) using C320 bitumen and PET-modified C320 binders (D)

RESULTS AND DISCUSSION

DSR RESULTS: UNAGED BINDER (OB) CONDITION

Figures 2 and 3 show the effect of different contents of waste PET modified binders through various testing temperatures on complex shear modulus (G*) and phase angle (δ). It can be seen that the addition of PET results

in increasing the complex shear modulus and reducing values of the phase angle. At the testing temperatures of 50 °C–70 °C, the modified binder tends to be substantially elastic materials, consequently, better rutting resistance is obtained. Results in Figures 2 and 3 argue that PET modified-bitumen samples were characterised with a notable rise in complex shear modulus (stiffness modulus), associated with a minor decrease in elasticity modulus and

the phase angle. This trend indicates that integration of the PET modifier results in an improvement in the elastic behaviour of the modified binder, and this phenomenon would be possibly connected to compatibility and swelling of the PET-modifier in asphalt binder (Leng et al. 2018).

Another reason might be related to the explanation of the current results of Figure 3 is the variety of physical and chemical properties of PET plastic and C320 asphalt. Generally, these properties are affected during the high shear mixing of plastic-bitumen blending and interaction process, which could result in changes in plastic-bitumenbinder particles dimension. As such, the engineering properties of the modified binder would be improved, as can be seen in Figure 4. The level of elasticity increases by extending the content of PET polymer. The increase in elasticity is due to the arrangement of molecules and the bonds with each other. In particular, when the temperature to the existence of cohesion is increased, or gaps between the chains are linked to each other, hence producing a more beneficial condition (Amir et al. 2019).

The phase angle (δ) describes the transformation between viscous-elastic behaviours of bitumen binders. Binders that become more viscous have a higher phase angle. Comparatively, lower values of phase angle correspond with higher elasticity. From the above results, it can be seen that the optimum PET content is 6%. This is shown through better rutting resistance as the PET modified binders improved the stiffness and viscose-elastic behaviour of the asphalt. As a result, binder rheologicalmechanical properties improved during the plastic polymer-bitumen phase separation and interaction. During the mixing process of bitumen and plastic polymer, the



FIGURE 2. Complex shear modulus vs. waste PET percentage



FIGURE 3. Phase angle vs. waste PET percentage

multi-stage arrangement of these interaction stages will occur, and viscosity will increase as a result of the strong bond of the polymer-bitumen interaction phase.

DSR RESULTS AFTER RTFOT AGEING

As PET modified bitumen is considered a new application in C320 Australian bitumen, it required a crucial need to examine the ageing behaviour during construction using the DSR and RTFOT tests. The results after RTFOT are shown in Figures 4 and 5, illustrating the impact of various PET content through different testing temperatures of 50 °C -70 °C on stiffness, elasticity, and rutting resistance. Figure 3 shows that adding waste PET increases the complex

shear modulus at all temperatures, like 50 °C, 58 °C, 60 °C, 64 °C, and 70 °C. Thus, it proves that the PET modified bitumen has improved stiffness, better durability, and ageing resistance than an unmodified binder.

This trend has been confirmed by the results of the phase angle, as displayed in Figure 5. The phase angle of the 6-8% PET modified bitumen shows an obvious decrease as compared to C320 (0% PET). This, in turn, results in a successful improvement in the elasticity of the modified bitumen binder. Consequently, improving the elasticity and stiffness of the modified binder, results to unenhanced resistance to deformation and displacement after subjected to oxidative ageing.

Figure 6 shows a promising application of PET modified C320 bitumen as it yields an indication of how



FIGURE 4. Complex shear modulus vs. waste PET % after RTFOT ageing



FIGURE 5. Phase angle vs. waste PET % after RTFOT ageing



FIGURE 6. Rutting Factor and ageing index at 64°C after RTFOT

the asphalt has less sensitivity to thermo-oxidative ageing and results in the incrementation in rutting factor by about 50% after RTFOT as compared to G*/ sin δ of unaged binders. The rutting resistance of all PET modified bitumen samples display a higher rutting factor at various temperatures of 50 °C-70 °C, as shown in Figure 6, with a better ageing index at 64 °C. As such, the PET modified binder will have a lower thermo-oxidative ageing, less hardening, and better bitumen-aggregate contact and bonding. As a result, the asphalt mix is less susceptible to cracking and deformation.

DSR RESULTS AFTER PAV AGEING

According to the report by The Strategic Highway Research Program (SHRP), a peak value of 5 000 kPa is recommended for the fatigue factor ($G^* \sin(\delta)$) and recommended low values are high indexes of fatigue properties. Figure 7 displays the fatigue factor, (G* sin (δ)), values of the C320 asphalt and different content of 4%-8% waste PET modified binders after (PAV) tests, were measured using (DSR) at the temperature of 25 °C- 40 °C. In general, the 8% PET content shows significant improvement to fatigue life of modified and results in fewer values of fatigue factor G* $\sin(\delta)$ of the plastic-modified binders. As shown in Figure 7, $G^* \sin(\delta)$, the modified bitumen is lower than the C320 unmodified bitumen at all temperatures of 25 °C-40 °C, indicating that PET polymer enhances the fatigue resistance of bitumen as a result of improving the PET polymerbitumen-phase interaction. At 25 °C, the fatigue factor was 4 200 kPa for C320 unmodified binder, and 2 100 kPa for 8% PET content modified binder.

According to a study by Leng et al. (2018), mixing PET and bitumen significantly impacts the fatigue properties in ways similar to commercial agents. This phenomenon is based on the fact that the chemical properties of PET organic compounds could possibly be changed by altering the number and position in the molecule system. The result show a significant consequence in the strength of asphalt, and as a result, led to the formation of molecular structures of PET-modifiedbitumen with increased tensile strength and elastic response which increases the fatigue resistance.

Figure 8 displays the fatigue properties before and after PAV ageing at a low temperature of 25 °C with an ageing index. It seems that with increasing the PET content in binder modification, the binder becomes stiff and can absorb the energy of traffic, resulting in longer fatigue life and less ageing index. This result shows that the ageing index of plastic modified asphalt is substantially less than C320 unmodified bitumen, meaning that the PET-modifiedbitumen would enhance the thermos oxidative ageing resistance of asphalt. This consequence, to some extent, would be described by the change of bitumen's chemical components during the ageing process that increases the asphaltene, which in turn is responsible for increasing the binder viscosity (Ali et al. 2013). Therefore, the polymerbitumen chains elongation increases at PET 8% and is working as a reinforced agent to resist shear force and reduce the fatigue cracking of bitumen.



FIGURE 7. Fatigue life vs plastic content after PAV



FIGURE 8. Ageing index at 25°C for unaged binders and after PAV ageing

CONCLUSIONS

The study investigates the impact of using local waste plastic as a sustainable modifier in C320 Australian bitumen. The remarkable findings of laboratory tests are below:

1. According to DSR results, samples modified with the waste PET show better performance in terms of improving the complex shear modulus and reducing the bitumen's susceptibility to deformation at high temperatures.

2. The RTFOT results specify that PET-binders had a low ageing index, high stiffness, better elasticity with a high rutting factor. Therefore, PET-modified bitumen binders show an excellent ability to resist ageing during construction and have better durability for long term services.

3. Results of PAV display that most PET modified asphalt samples have better fatigue resistance, lower ageing index, and longer fatigue life as compared to unmodified C320 bitumen.

4. Future tests should conduct various mixing conditions

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and quantify the reinforcement capability of PET modified asphalt mixtures.

DECLARATION OF COMPETING INTEREST

None.

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