



The Utilisation of Polyethylene Waste Pyrolytic Wax as an Additive to Enhance the Performance of Virgin and Polypropylene-Modified Bitumen

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Abstract

Plastic addition to bitumen increases the stiffness of the binder, reduces its workability due to the increase in viscosity, and causes storage stability issues. Thus, plastic pre-treatment methods are needed. This research aims to address these shortcomings by developing a wax derived from the pyrolysis of polyethylene (PE) waste and investigating its efficacy as an additive in virgin bitumen and bitumen modified with recycled polypropylene (PP). Penetration, softening point, segregation, stress ratio, consistency, stiffness, and viscosity of the modified bitumen were measured. The interior structure and chemical changes of the proposed binders were also studied using a fluorescence microscope and Fourier-transform infrared spectroscopy (FTIR). When virgin bitumen is modified with 7% PE wax, its softening point increases (+15%), its viscosity is reduced (−27%), and no segregation issue is observed. It also presented a stiffness increase (+32%) and better resistance to hot climates. Adding 7% of PE wax to bitumen modified with 5% PP reduces its viscosity by 70% and segregation by 26% while increasing its stiffness and rutting resistance in comparison to the bitumen solely modified with PP. These results reveal that, in addition to enhancing bitumen properties, the proposed substance reduces compaction and mixing energy requirements.

Keywords Pyrolysis · Plastic waste · Hot mix additives · PE · PP · Bitumen modified

Abbreviations

ARRB	Australian Road Research Board	PWMB	Bitumen modified with pyrolytic wax and polymer
C320	Australian bitumen type C320	PP	Polypropylene
DSR	Dynamic shear rheometer	PPC	Plastic pyrolytic char
FM	Fluorescent microscope	PPO	Plastic pyrolytic oil
FTIR	Fourier-transform infrared spectroscopy	PPW	Plastic pyrolytic wax
HMA	Hot mix asphalt	PVC	Polyvinyl chloride
LCA	Life cycle analysis	WMA	Warm-mix asphalt
NATA	National Association Testing Authorities	WMB	Pyrolytic wax-modified bitumen
NCHRP	National Cooperative Highway Research Program	ZSV	Zero shear viscosity
PET	Polyethylene terephthalate		
PEX	Cross-linked polyethylene		
PI	Penetration index		
PMB	Plastic-modified bitumen		

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1 Introduction

The use of plastic waste as an additive to pavements in road construction has recently gained interest [1, 2]. Plastics are added to pavement through two methods: dry and wet mixing. In dry mixing, plastic is added to hot aggregate and is mixed with the hot bitumen. In the wet technique, plastic is added and combined with the hot bitumen before adding the aggregates. Commonly, both methods produce stiffer asphalt mixes and pavements more resistant to rutting [1, 3]. The advantages of the dry method are that it allows for

high quantities of plastic additives, does not require complex equipment, and is easier to implement [4]. However, the even distribution of plastic in the asphalt mixture is not guaranteed, and depending on the plastic type, melting point, and particle size, it could deteriorate the mixture's cohesion [5]. Alternatively, the benefit of the wet technique is that it directly modifies the bitumen. By doing so, this mixing technique—compared to the dry approach—is more effective in modifying the final properties of the asphalt mixture [3]. Nevertheless, it also presents two major issues when the plastic addition is excessively high: high viscosity and segregation issues (storage stability). Bitumen with high viscosities requires higher energy inputs for the appropriate mixing with the aggregates.

Segregation occurs when the plastic separates from the bitumen after it has been blended. To remedy this difficulty, the plastic must have an appropriate chemical affinity with the bitumen and their densities must be comparable [5, 6], which is not always attainable due to the inherent qualities of plastic waste. To overcome this problem, plastic pretreatments are needed so that the final plastic density and affinity are comparable to bitumen.

The prevailing pre-treatment methods in the literature can be classified into three categories: chemical reaction, electron irradiation, and pyrolysis. Chemical reaction uses a chemical to alter the plastic composition and improve its affinity with the bitumen. The chemicals used for the modification of the plastic vary, for example, Padhan and Gupta [7] employed ethanolamine to modify PET, while Leng et al. [8] used an aminolysis process. In terms of other plastic types, Ahmedzade et al. [9] modified PP through the addition of 1, 2, 4-trichlorobenzene, and even PVC, a plastic that is dangerous due to the toxic release when heated, was also chemically modified and evaluated as a bitumen modifier [10]. In general, although chemical modification preserves the common plastic benefits on bitumen, such as better rutting resistance, it still deteriorates the viscosity. Among these studies, only Behl et al. [10] assessed the storage stability property. They found that although this property is degraded, its value is still within the permissible range of the bitumen standards. A similar viscosity response was observed when irradiation was employed [11, 12].

Pyrolysis is the third method employed to produce additives used to modify bitumen. These additives are usually plastic pyrolytic oil (PPO), plastic pyrolytic wax (PPW), and plastic pyrolytic char (PPC). Al-Hadidy tested bitumen modified with plastic pyrolytic products [13, 14]. Regardless of the plastic type, Al-Hadidy's results were consistent with the other pre-treatment methods: better rutting resistance but higher viscosity. In contrast, when PPO is used as an additive, the effect is the opposite: the softening point decreases and deteriorates the bitumen's rutting resistance. Avsenik and Tušar [15] observed that adding PPO to the bitumen

decreased the softening point and increased the penetration, which involved permanent deformation risks and poor performance in hot climates. To solve this rutting issue, some authors have further modified the PPO through the distillation and extraction of its heavy fraction. This extra step in the plastic pre-treatment has favoured the creation of a bitumen resistant to hot climates with a higher viscosity [16, 17].

Interestingly, Ling et al. [18], who combined PPO and PPW, improved bitumen resistance in cold climates. They even concluded that this modifier could be used in warm-mix asphalt. Warm-mix asphalts require less temperature, approximately 20 °C less to manufacture and pave than hot asphalt mix. These positive results of PPW are not limited to the cases where it is combined with PPO. Mohammed and Hussein [19] and Shang et al. [20], who added the wax fraction exclusively, noticed a decrease in the viscosity, better rutting resistance, and an acceptable segregation value. Due to the usual trade-off observed in asphalt mixture stiffness, it is interesting to note that these two results appear conflicting. Asphalt mixtures that are stiffer are more resistant to rutting, but their susceptibility to cracking and cold-temperature cracking decreases. When the asphalt mixture's stiffness is decreased, the opposite occurs. Thus, although PPO and PPW could produce an asphalt mixture that requires less temperature, the final stiffness of a mixture modified with these components will depend on the qualities of the pyrolytic outcome and, consequently, on the pyrolysed material.

Although the pre-treatment methods could facilitate plastic usage in bitumen, some concerns still exist related to the increase in cost and the requirement of complex equipment. Nevertheless, the use of PPW could be the most attractive because the other components not used from plastic pyrolysis (oil and gas) can be used as an energy source, reducing the overall cost and environmental burden. With this in mind, the present article aims to formulate a suitable PPW, evaluate its effect as an additive to bitumen and polymer-modified bitumen, and identify an optimal bitumen/wax/polymer ratio. The plastics used are PE and PP because they makeup more than 50% of total plastic waste globally; in 2015, 32% were PE and 18% were PP [21].

To achieve the proposed aim, pyrolytic wax will be produced from PE and PP, and it will be mixed with virgin and polymer-modified bitumen. After formulating multiple samples with different percentages of additives, basic properties such as softening point, penetration, and segregation will be measured and compared. Through the measurements of mixing temperature and compaction, we will evaluate if the wax obtained from the pyrolysis could reduce the energy requirements for asphalt mixture formulation. In the case where the wax is mixed with the polymer, we hypothesise that it could reduce viscosity and segregation issues. Some researchers have previously studied PE wax as a suitable pavement additive [20, 22, 23]; nevertheless, these studies

were based on commercial waxes instead of pyrolytic. In addition, although other studies have also tested PPWs, none focused particularly on PP or PE. Ling et al. [18], for instance, tested the wax produced from PET mixed with oil from crumb rubber. Similarly, Mohammed and Hussein [19] exclusively evaluated the wax produced from PET. In addition, Shang et al. [20] focused on the wax produced by cross-linked polyethylene (PEX) and styrene-butadiene-styrene (SBS) as a bitumen modifier. Although these studies provided a sense of what could be expected by combining pyrolytic wax with bitumen, they did not specifically evaluate the bitumen's reactions and responses towards PP and PE in the waxy form. In addition, they did not evaluate the combined effect of adding pyrolytic wax to bitumen that has been modified with recycled plastic. The latter two aspects constitute the novel contribution of the present study.

2 Materials and Methods

Figure 1 summarises the procedure to be followed for the evaluation of the PPW as a binder additive. The first phase, plastic pyrolysis, will encompass the pyrolysis of the plastics and the selection of the best pyrolytic outcome based on the FTIR and the consistency of the material. During this screening, in addition to choosing the most suitable wax additive, the optimal pyrolysis configuration that yields the most suitable PPW is also determined. Optimal wax will be used in “Binder formulation and initial tests”. At this stage, modified bitumen will be formulated with plastic, PPW, or a combination of both at different percentages. Softening point, penetration, and segregation tests will be conducted on the formulated binders to select the most

promising mixtures. For the advanced tests, the optimal binders screened in the previous stage will be evaluated in more advanced rheological tests and chemical and microscopic characterisation procedures using the FTIR and the fluorescent microscope. Among these advanced tests, the viscosity at 135 °C and 165 °C, stress ratio, consistency, and stiffness were evaluated by an external NATA (National Association Testing Authorities) laboratory. One sample of each screened bitumen type was sent to the laboratory for analysis. In Australia, laboratories with NATA accreditation are guaranteed to provide services of high quality, safety, and reliability.

2.1 Plastic Pyrolysis

Three plastic compositions were evaluated during the pyrolysis of the plastics. The first was made entirely of HDPE (high-density polyethylene) (100% HDPE), the second entirely of PP (100% PP), and the third was a mix of HDPE and PP (44%:56% proportion). This percentage represents the equivalent consumption of PP and PE in Australia from 2017 to 2018 [24]. Some properties of these plastic types are presented in Table 1(a, b) and Fig. 2a, b. The referred reactor consisted of a heating mantle and an enclosed system of a 1 L flask connected to a condenser (Fig. 3). Instead of using inert gas to displace the oxygen, a vacuum system was applied within the enclosed system. In this pyrolytic stage, two parameters were evaluated: residence time (30 and 45 min) and temperature (350, 400, and 415 °C), to determine and maximise the wax outcome. The yield for each combination was calculated. After the pyrolysis had been completed, oil and wax outputs were collected and weighted for yield measurements. While the pyrolytic oil

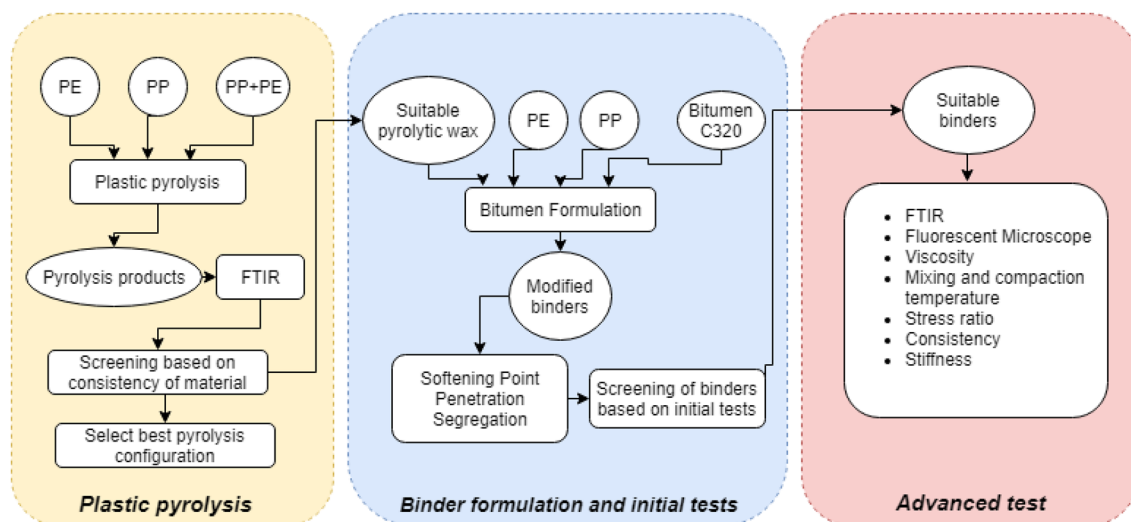


Fig. 1 Experimental procedure

Table 1 Plastic properties

Property	Result
HDPE plastic lids (pyrolysis) (a)	
Maximum particle size	5 mm
Density at room temperature	0.9165 g/cm ³
Melting point	135 °C
PP Plastic cups (pyrolysis) (b)	
Maximum particle size	5 mm
Density	0.8917 g/cm ³
Melting point	141 °C
Recycled HDPE (bitumen polymer modifier) (c)	
Maximum particle size	5 mm
Density	0.9209 g/cm ³
Melting point	125 °C
Recycled PP (bitumen polymer modifier) (d)	
Maximum particle size	5 mm
Density	0.9857 g/cm ³
Melting point	164 °C

was condensed from the pyrolysis gases in a separate flask, the wax was collected from the reactor's bottom. In the case of the PP, instead of wax, heavy fraction oil was deposited at the bottom of the reactor.

Only the waxes produced by pyrolyzing 100% HDPE will be studied further as potential bitumen additives. PP pyrolytic oil has a flashpoint of 30 °C [25], which may jeopardise the safe handling of the resulting bitumen. According to Australian standards, the flash point of the pyrolytic oil is unacceptable, even to be used for reducing residual bitumen viscosity, where a minimum flash point of 61.5 °C is required [26]. Furthermore, due to the possibility of low flashpoint bitumen formation due to the PP oily fraction, the wax produced by pyrolyzing a combination of PP and HDPE (44%:56% proportion) was also excluded.

The chemical components of the pyrolysis outcomes were analysed with the FTIR technique. One sample per pyrolysis output was evaluated. Perkin-Elmer Spectrum Two FTIR instruments were utilised. Each of the samples was placed directly on the FTIR crystal. After each measurement, the crystal was cleaned with a natural solvent, and between

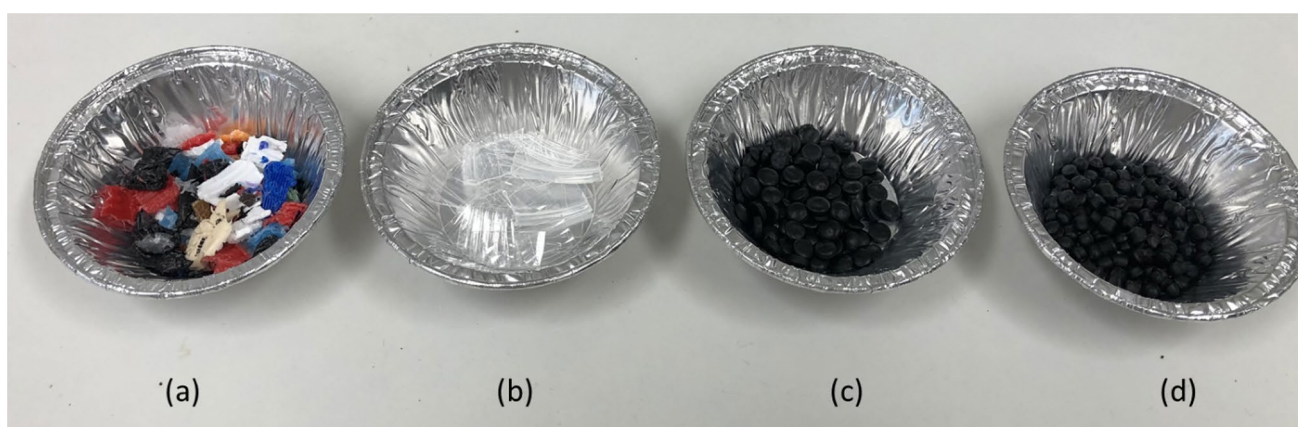
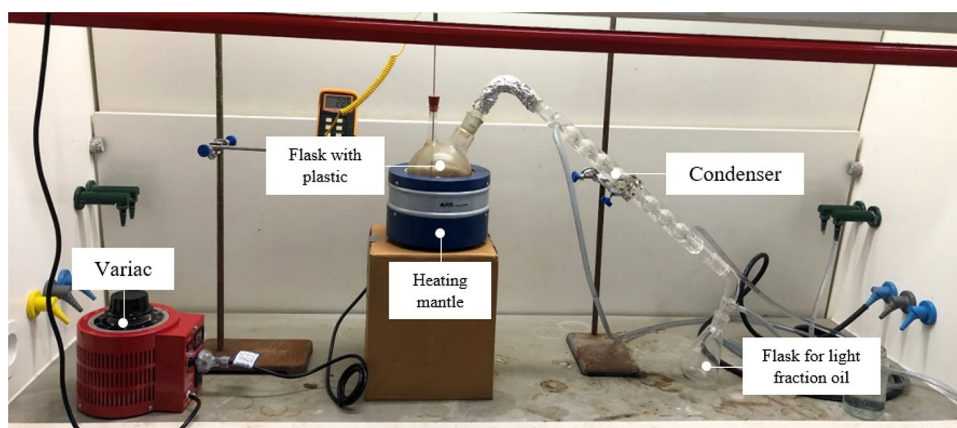
**Fig. 2** a HDPE plastic lids, b PP plastic cups, c PE reclaimed pellets, d PP reclaimed pellets**Fig. 3** Pyrolytic reactor

Table 2 Bitumen C320 properties

Property	Standard	Result
Viscosity at 135 °C	AS 2341.4	0.59 Pa.s
Penetration at 25 °C	AS 2341.26	4.56 mm
Softening point	AS 2341.18	46.27 °C
Density at 15 °C	AS 2341.7	1.05 g/cm ³
Flash point	AS 2341.14	360 °C

measurements, background measurements were taken. The wavenumbers studied ranged from 450 to 4000 cm⁻¹. Melting points were also measured by following the standard ASTM D87-04 [27]. Notably, although the by-products of pyrolyzing the three plastic compositions were analysed by FTIR, only the wax produced from the 100% HDPE composition was used as an additive in the subsequent bitumen tests.

2.2 Binder Formulation and Initial Tests

2.2.1 Binder Formulation

The bitumen used to prepare the samples was the Australian C320 (Table 2). This bitumen was obtained from Puma Energy and Austek in Queensland, Australia. According to the Australian Standard, C320 bitumen is used for base and wearing courses, and it is often employed in the formulation of polymer-modified bitumen [28, 29]. Three forms of modified bitumen were proposed: plastic-modified bitumen (PMB), pyrolytic wax-modified bitumen (WMB), and bitumen modified with pyrolytic wax and polymer (PWMB). HDPE and PP were used in the PMB and PWMB formulations. Some properties of these plastics can be found in Table 1 and Fig. 2. The company Resitech, in Brisbane, Australia, supplied these plastic resins with a maximum size of 5 mm. For producing the PMBs, plastic was added to hot bitumen at 180 °C and stirred at 900 RPM for one hour. The selection of this mixing protocol was based on previous studies of bitumen modified with plastic waste [30–32]. The WMB was formulated by mixing the wax obtained after the pyrolysis stage with hot bitumen. A special emphasis should be made on the temperature and time during mixing, as high temperatures and oxygen can accelerate the ageing of the binder [33]. In the case of PMB, it was not possible to reduce the temperature or the mixing time since this would result in a non-homogeneous bitumen. However, in the case of WMB, it was observed that a mixing temperature of 150 °C and a mixing time of 30 min were sufficient to synthesise a suitable binder. This was possible because of the low melting point of the wax and its easy dispersion in the bitumen during mixing. The selection of this mixing setting was also based

on the work of Zhang et al. [34], who evaluated the effect of commercial PE waxes on bitumen. On the basis of the findings of Hunter et al. [35], it is possible to quantify the ageing effect of these two protocol temperatures. According to Hunter et al. [35], after reaching 100 °C, the ageing rate of bitumen doubles every 10 °C. Consequently, the ageing rate of mixing at 180 °C is estimated to be eight times that of mixing at 150 °C. Regarding the effect of mixing time, there is no estimate in the literature. However, one can argue that mixing the bitumen for a longer time will increase the binder surface area that is in contact with oxygen. As a result, mixing bitumen for one hour rather than 30 min may result in a faster ageing rate.

The PWMB formulation included the addition of pyrolytic wax and recycled plastic. The mixing conditions of the PWMB were comparable to those of the PMB preparation, and the addition sequence was wax followed by plastic. Table 3 summarises the formulation of the samples and the percentage of addition of each component over bitumen weight. The initial percentages considered for the PMB were 5, 7, and 10%. However, the 10% was later removed because it did not yield a stable modified bitumen. Regarding the WMB, the wax percentage was set at 2, 3, 5, 7, and 10%, which is based on previous studies of similar materials [13, 17–19, 36]. In the case of the PWMB, the plastic addition remained constant at 5% while the wax component varied (2, 3, 5, 7, and 10%). This constant plastic content was chosen based on previous reviews [1, 37–39] and an initial screening of the PMBs.

Table 3 Samples' formulation

Sample name	Sample type	Percentage of addition over bitumen weight (%)		
		Recycled PP	Recycled PE	Pyrolytic wax
C320	Virgin bitumen	–	–	–
PP5	PMB	5	–	–
PP7	PMB	7	–	–
PE5	PMB	–	5	–
PE7	PMB	–	7	–
W2	WMB	–	–	2
W3	WMB	–	–	3
W5	WMB	–	–	5
W7	WMB	–	–	7
W10	WMB	–	–	10
PW2	PWMB	5	–	2
PW3	PWMB	5	–	3
PW5	PWMB	5	–	5
PW7	PWMB	5	–	7
PW10	PWMB	5	–	10

2.2.2 Initial Tests

The penetration and softening points were measured in accordance with Australian Standards AS2341.12 [40] and AS2341.18 [41]. Although the penetration measurement of modified binders does not allow for their entire characterisation, it nonetheless provides a basic overview of their consistency [35]. In addition, it has been used regularly in prior papers examining binders modified with plastic waste. Alternatively, the softening point is a broad assessment of bitumen's susceptibility to high temperatures; a higher softening point typically indicates a greater resistance to hot conditions [42]. While the penetration measurement is not included in the Australian polymer-modified framework (AGPT-T190-19), the softening point remains valid in this standard and is also the primary test applied for the segregation test method. To increase the statistical reliability of the final estimations, three different samples per bitumen type were measured.

The segregation was also evaluated and implemented as described in the standard AGPT-T108-06 [43]. The bitumen sample was poured into an aluminium container and placed in an oven at 180 °C for 48 h. The sample was then removed and cooled to room temperature before being cut vertically. A photograph of the exposed side of these samples was taken. Both halves were cut horizontally, and the two top quarters were merged, as were the bottom quarters, yielding two subsamples. The softening point of these subsamples was measured, and the final segregation is calculated following Eq. (1). Two distinct samples per bitumen type were analysed. According to Austroads, regardless of the PMB, the maximum permitted limit of the segregation property (S) is 8%:

$$S = \frac{200 \times (T_t - T_b)}{T_t + T_b}, \quad (1)$$

where S is the segregation (%), T_t is the softening point top section (°C), and T_b is the softening point bottom section (°C).

2.3 Advanced Tests

2.3.1 Fluorescent Microscope and Fourier Transform Infrared Spectroscopy Analysis

The fluorescent microscope (FM) was used to examine the additive dispersion in the samples. For a better understanding of the phase separation impact, FM was also used to assess the top and bottom samples used during the segregation test. In order to prepare the samples, a droplet of hot bitumen was deposited on a glass slice and sandwiched between two microscope slides. After the droplet had

reached room temperature, the assembly was placed in a 135 °C oven for 5 min. Clamps were employed to keep the samples in place while they were heated. After 5 min, the pieces were removed from the oven, allowed to cool, and then stored in the freezer for further examination under the fluorescence microscope. One sample per screened bitumen type was evaluated under the FM. One sample per screened bitumen type was evaluated under the FM.

2.3.2 Viscosity and Mixing/Compaction Temperatures

Samples were tested for viscosity in accordance with the Australian standard AS2341.4 [44], which is comparable to the American standard AASHTO T316 [45]. The instrument utilised was the Brookfield LV (low viscosity) and its corresponding thermosel system. The type of spindle was SC4-31. The viscosity was measured at temperatures of 135 and 165 degrees Celsius and at a shear rate of 34 s⁻¹. Bitumen, particularly polymer-modified bitumen, must have a viscosity of less than 3 Pa.s at 165 °C in order to be easily pumped, transported, and mixed with aggregates at mixing temperatures [46]. The equiviscous approach, explained in the ASTM D 2493 [47], was utilised to deduce the mixing and compaction temperatures. A consensus exists, however, that this approach is not accurate for polymer-modified binders and binders modified with wax, since it tends to overestimate the temperatures obtained. The National Cooperative Highway Research Program (NCHRP) has stated that the equiviscous approach may not apply to a large variety of modified binders. Other tests, such as Zero Shear Viscosity (ZSV), simplified ZSV, the high shear rate approach, and the flow behaviour method, have produced results that are less exaggerated and, arguably, more accurate [48]. Even though each test yields unique results, they all, including the equiviscous method, share a common trait: bitumen with lower viscosity values requires a lower temperature for compaction and mixing than binders with a high viscosity. This tendency has been observed in polymer-modified binders [49], bitumen treated with commercial waxes [50], binders modified with PE wax [51], binders modified with pyrolytic char [52] and even in asphalt mixtures [34]. Although these tests could provide a more precise measurement, they are in line with the trend observed using the equiviscous method, as demonstrated in previous articles [48, 50, 52, 53]. Considering this fact and the absence of a standard for the measurement of compaction and mixing temperatures among binders modified, we consider the use of the equiviscous procedure a good proxy technique to determine these two critical temperatures. The viscosity measurements at 135 °C and 165 °C were taken to deduce the mixing and compacting temperatures, which correspond to viscosities of 0.17 Pa.s and 0.28 Pa.s, respectively [54, 55].

2.3.3 Stress Ratio

The test employed to assess the binder's resistance to cold temperatures was the stress ratio through the dynamic shear rheometer (DSR). The Australian specification for this procedure was AGPT/T125 [56]. The procedure's temperature is set to 10 °C, and the DSR is programmed to impart rotational shear on the sample at a constant rate of 0.0075 s⁻¹. The stress and strains are then measured, and the stress ratio is calculated as the stress at the 10 strain divided by the stress at 3 strain. According to Austroads [57], there is a reasonable link between the stress ratio measurement and the fatigue life of mixes at 10 °C. The measurement has no threshold in the general Australian PMB framework (AGPT/T190) [58]; however, it must be reported. In general, binders with greater stress ratios correlate to longer asphalt mixes' fatigue life at cold temperatures.

2.3.4 Consistency at 6%

The evaluation of the rutting risk of the proposed binders is done through the measurement of the consistency at 6% of the binder at 60 °C. This test was done following the Australian standard AGPT/T121 [59]. The instrument employed is the ARRB (Australian Road Research Board) elastometer, which applies an axial strain to an annular sample that has been sandwiched between two cylinders. The strain rate is constant at 0.1 s⁻¹, and the test temperature is 60 °C. The consistency at 6% is the stress recorded at a 0.06 strain (Pa) divided by the actual strain rate (s⁻¹). As demonstrated by Urquhart et al. [60], the consistency of a particular binder measurement is a good predictor of the wheel tracking results of an asphalt mixture formulated with the mentioned binder. The higher the consistency, the lower is the rutting damage in asphalt mixtures.

2.3.5 Stiffness

The stiffness measurement, like the consistency measurement, is taken from the ARBB elastometer at a constant strain rate of 0.1 s⁻¹, but at a reduced temperature of 25 °C. According to AGPT/T121, stiffness is defined as the ratio of peak stress to strain. The peak stress is the force at a certain strain or breaking point predefined in the ARRB elastometer divided by the area of the annular sample [59]. Since this test is performed at 25 degrees Celsius, it provides a general indication of the binders' proclivity to produce fatigue cracking in asphalt mixes. This sort of cracking arises as a result of the asphalt pavement layer's continual deflection and recovery, and pavements are more vulnerable to this type of damage when the binder is stiffer [61].

3 Results and Discussion

3.1 Plastic Pyrolysis

The PP and HDPE pyrolytic products were recovered after pyrolysis. The final HDPE outcomes were wax and light fraction oil. Conversely, while the PP produced a light fraction oil too, it created a heavy fraction oil (HFO) rather than a wax. The resultant components of the pyrolyzed PP and HDPE combined were the same as for HDPE alone: light fraction oil and wax. Figure 4 depicts the FTIR results for each pyrolytic component and the unmodified C320 bitumen. All these pyrolytic components were obtained at a temperature of 400 °C with a residence time of 30 min. In general, it is possible to divide these results into waxy and oily materials. This division is detected within the fingerprint region (500–1500 cm⁻¹). Alternatively, both groups present similar spectra peaks in the waves length of the aliphatic compounds of the methyl (CH₃) and methylene (CH₂), 1485–1500 cm⁻¹ and 2800–2946 cm⁻¹, and a small peak in the aromatic ring stretch (C=C) corresponding to the wavelength 1637–1547 cm⁻¹. When compared to C320 bitumen in the fingerprint area, there is a clear difference between the pyrolytic components and the neat bitumen. Changes outside this zone are most noticeable near the wavelength areas of 1000 cm⁻¹ and 1600 cm⁻¹. However, outside of the fingerprint area, all of these compounds and the C320 bitumen exhibit the same major peaks. This suggests that their functional groups are similar, and when combined with bitumen, they could display good affinity within the binder.

As stated in the methods, the only material studied further from this point forward will be the wax produced by the pyrolysis of 100% HDPE. Figure 5 displays the FTIR analysis of HDPE wax under different pyrolysis conditions. As observed, the temperature and residence time during pyrolysis have little effect on the pyrolytic wax's chemical makeup. Figure 5 also depicts the FTIR profile of the unaltered polyethylene, which reveals that the HDPE experienced a chemical transformation after pyrolysis (as observed in the fingerprint region of the figure). Since there was no noticeable variation in the chemical composition of the pyrolysis setups, we opted to utilise the pyrolysis configuration that maximises the wax production. This configuration was at 350 °C for 30 min, which yields 91% of PPW. The wax obtained in this configuration displayed a melting point of 102 °C. It is essential to note, however, that these pyrolytic parameters might be modified in the future to alter the yield and, potentially, make the process more economically viable by utilising the other pyrolytic products (light fraction oil and gas).

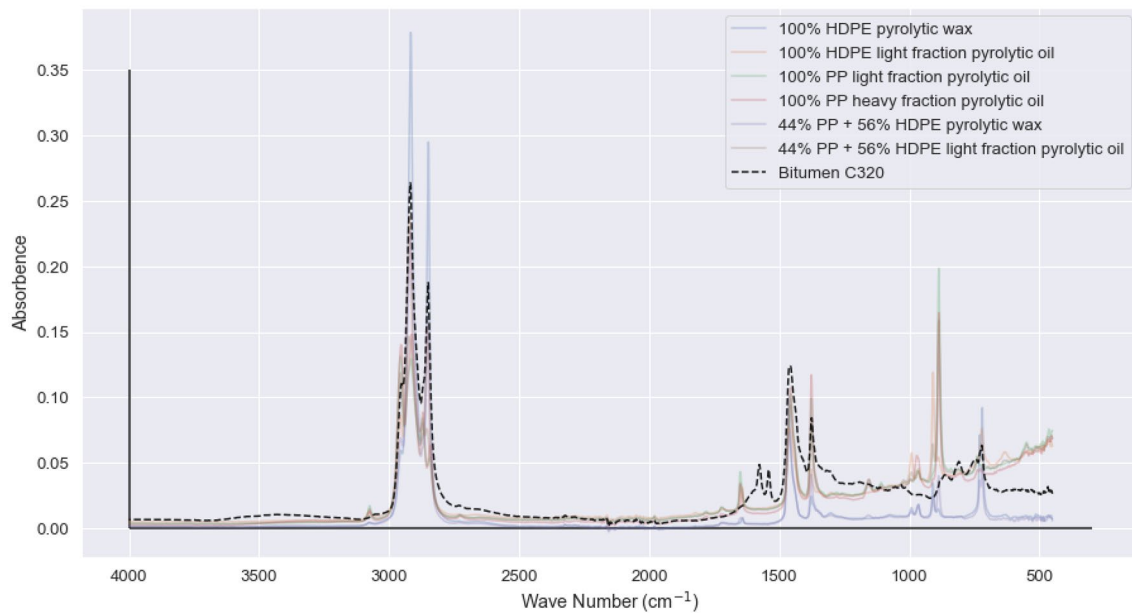


Fig. 4 FTIR for the different outcomes of plastic compositions pyrolysis

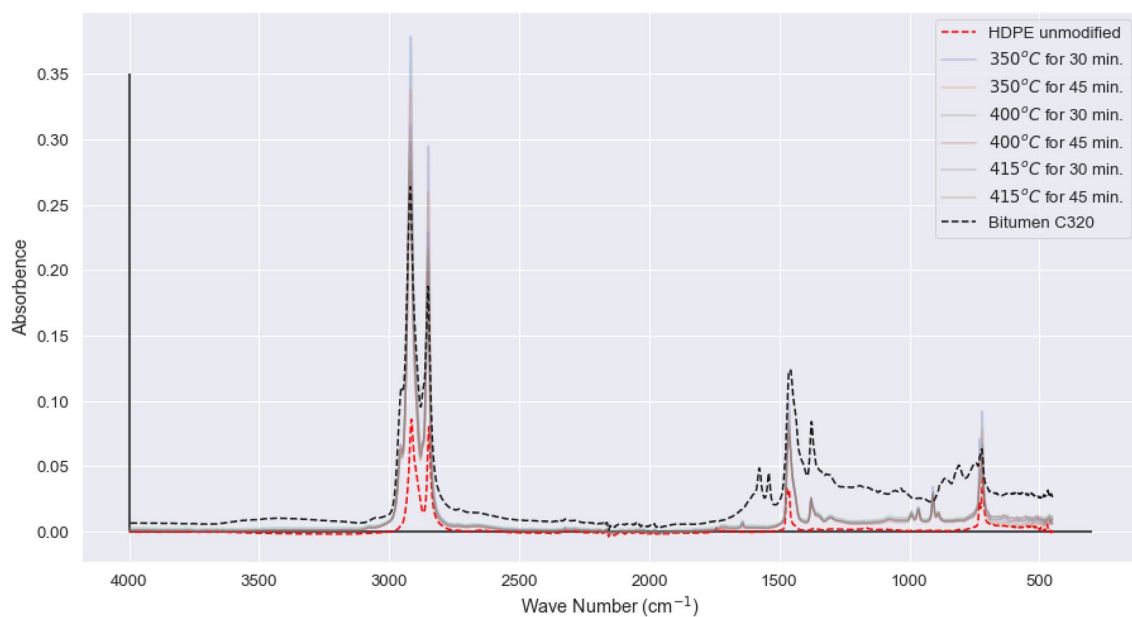


Fig. 5 FTIR analysis of HDPE wax for different residence times and temperature

3.2 Initial Bitumen Tests

3.2.1 Penetration

The addition of recycled PE and PP results in a decrease in the penetration property of the bitumen. Figure 6 presents the penetration for each of the bitumen types evaluated with their respective standard deviations. These effects were expected, given that

they had been observed in previous studies on binders enhanced with plastic [1, 62]. This is caused by the creation of a polymer-bitumen network structure, which is also the reason for the stiffening of the bitumen [63, 64]. Similarly, Brasileiro et al. [62] confirmed that PE has a greater influence on penetration reduction than PP. The above effect is likewise proportional to the amount of plastic added; the more plastic added, the greater the decrease in penetration.

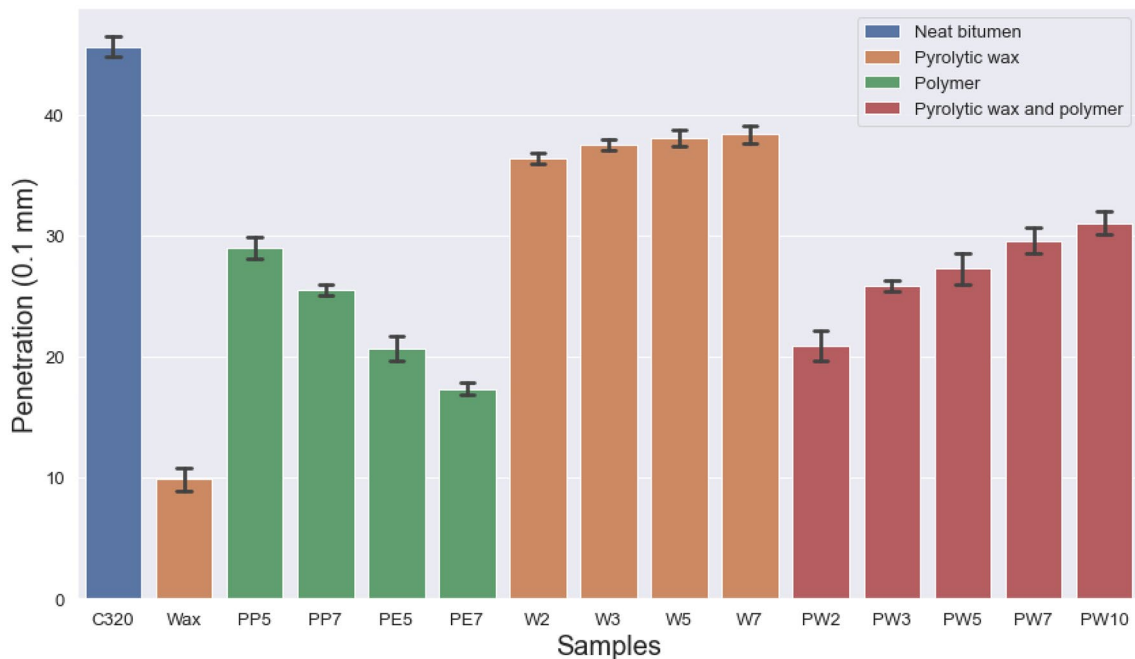


Fig. 6 Penetration of PMB, WMB, and PWMB

A one-way ANOVA revealed a statistically significant difference in the penetration of the majority of the sample groups ($F(14, 129) = [1067.18]$, $p = 5.75e-126$) (Annex A). Tukey's test was used to determine statistical differences between pair-sampling combinations. Except for PE5–PW2, PP5–PW7, PP7–PW3, and several samples with just wax modification (W2–W3, W3–W5, W3–W7, and W5–W7), the majority of the samples were statistically different ($p < 0.05$) (Annex B).

When compared to plain bitumen, Fig. 6 illustrates that PPW decreases penetration, confirming that the wax can augment the bitumen's hardness. Even a small addition of wax (W2) produced a bitumen with a significantly lower penetration than the C320 bitumen (Tukey HSD; $p < 0.001$). This early observation suggests that the PPW may provide the same benefits as plastics, albeit with less effect on the penetration than PP and PE. Commercial waxes, such as Montan wax and FT-paraffin [65, 66], and commercial PE waxes [67] have likewise shown a decrease in the penetration of binder, confirming that the findings are consistent with the nature of the substance.

In terms of the samples modified with polymer and wax, it is interesting to note that penetration increased with the increasing wax content in the polymer-modified bitumen. Nonetheless, because the combination of wax and polymer produces a binder with less penetration than the control bitumen, even when the wax addition is high (Tukey HSD [C320 vs. PW10]; $p < 0.001$), these combined additives can still produce a harder binder. Among previous studies

that evaluated the combined addition of polymer and wax, Rodríguez-Alloza et al. [68] observed a contradictory effect on the incremental addition of commercial waxes—higher wax quantities decrease the penetration—and this was also true for Shang et al. [20]. Factors concerning the internal structure of the waxes investigated, as well as their interaction with hot bitumen and polymers, could be creating this difference with the current results.

3.2.2 Softening Point

The softening point of each of the binders evaluated is depicted in Fig. 7 along with their respective standard deviations. A one-way ANOVA test found statistically significant differences across groups ($F(13, 76) = [1479.86]$, $p = 1.05e-85$) (Annex C). The only combinations with no statistically significant differences in softening points after the Tukey's test were PP5–PW2, PP5–PW3, PP5–W5, PW2–PW3, PW2–W5, PW3–W5, PW5–PW7, PW5–W7, and PW7–W7 (Annex D). The softening point has long been associated with bitumen's tolerance to hot temperatures. The effect of the polymer, as shown in Fig. 7, was expected because it has been widely reported in the literature that the inclusion of these additives increases the rutting resistance of the binder [1, 69]. The PE addition has a stronger effect on the softening point than the PP additive (Tukey HSD [PP5 vs. PE5] and [PP7 vs. PE7]; $p < 0.001$). Habib et al. [70] also found that PE has a greater effect than PP, and Brasileiro et al. [62], in their literature review, indicated that this effect is most

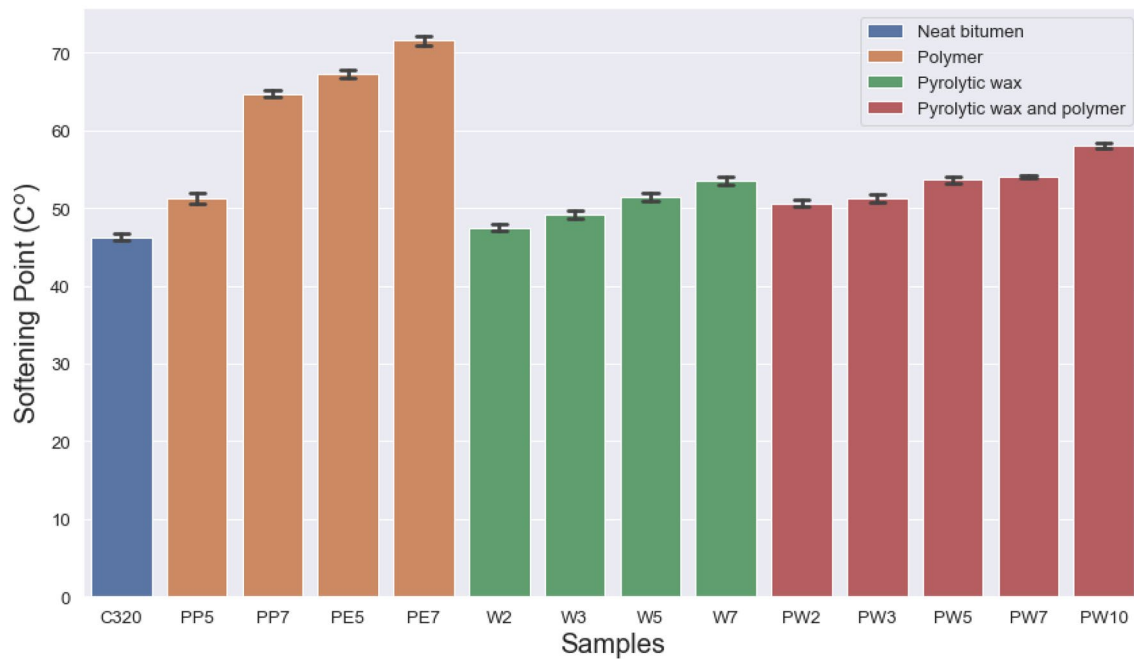


Fig. 7 Softening point of PMB, WMB, and PWMB

noticeable in binders including PP and PE at concentrations ranging from 1 to 9%.

As with the addition of polymer, the addition of wax raises the bitumen's softening point. Although the rise is not as significant as that of the polymer, it is enough to distinguish itself from the neat bitumen (Tukey HSD [C320 vs. W2]; $p = 0.0059$ —Tukey HSD [C320 vs. W3], [C320 vs. W5] and [C320 vs. W7]; $p < 0.001$). This result shows that HDPE wax could be used for hot-climate pavements. When adding wax to bitumen, the degree of wax crystallisation is a crucial consideration. This crystallisation occurs during the cooling of certain waxes, and it is a feature that should be investigated carefully. A high degree of crystallisation reduces the bitumen's resistance to plastic deformation and cracking. However, a tiny amount of crystallisation may be advantageous since the wax-modified bitumen becomes more viscous as the temperature falls below the wax's melting point [71]. This reduces the likelihood of permanent deformation [72]. In general, waxes are classified as macrocrystalline, microcrystalline, or non-crystalline. Microcrystalline wax produces smaller crystals that increase the viscosity of bitumen, whereas macrocrystalline wax produces larger crystal formations that are undesirable in the binder. In addition, microcrystalline waxes are preferred as binder additions because they dissolve more readily in bitumen and, usually, reduce mixing and compaction temperatures. Thus, it might be plausible to think that the wax formulated presents some degree of microcrystalline formation, as was also suggested by other articles [73, 74].

The effect found in bitumen modified with wax and polymer appears to be the sum of the effects of the individual additives' effects. The addition of 5% PP (PP5) resulted in an increased softening point of 51.3 °C, while the addition of 2% wax (PW2) resulted in a value of 50.63 °C, which is deemed generally equivalent (Tukey HSD [PP5 vs PW2]; $p = 0.5493$). Then, after the gradual addition of the wax, the softening point increased at the same rate as that observed in the binder modified solely with the wax. Two effects are observed when the wax and the polymer are included. The first one is provoked by the network created by the polymer, which creates an internal structure more resistant to hot temperatures. Then, with the wax addition, this internal structure is reinforced by the crystallinity of the wax, which is causing the progressive increase observed in Fig. 7.

3.2.3 Segregation

Figure 8 depicts the segregation property and standard deviations for each of the formulated samples. As expected, the binder treated with recycled plastic displayed high segregation, confirming prior research on PP and PE [69, 75]. Based on the Australian standards for PMB [58], the maximum segregation value is eight, and any bitumen with a higher value is considered unsuitable. As observed in Fig. 8, none of the bitumen modified with polymer complied with this limit. Nevertheless, it is still interesting to see that PP presents a lower impact than PE. This behaviour could be caused by the density difference between plastic types. According to

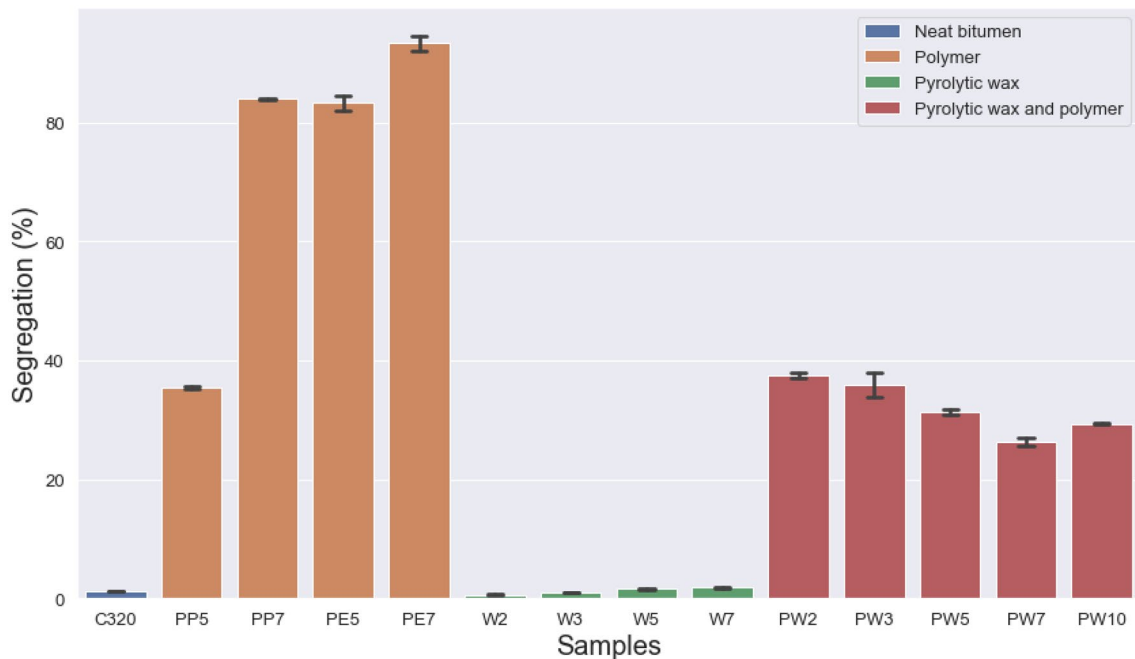


Fig. 8 Segregation values

some authors [76, 77], one of the major causes of segregation issues is the polymer-density difference, so because the PP bears a density closer to the bitumen's, it might be causing the observed lower effect on the final segregation value.

Regarding the pyrolytic wax, no segregation issues were observed, and their values were similar to the unmodified bitumen. Seven percent of addition (W7) featured the highest segregation, 1.79. This result is inconsistent with those obtained by Zhang et al. [34] who also studied bitumen modified with polyethylene wax; however, they found a higher segregation value (between three and 16). Interestingly, they also obtained negative segregation values, which means that the wax sank into the bitumen, rather than floating as happened in the present article. The discrepancies in results can be explained by the particularities of the bitumen and wax studied. Zhang et al. [34], for instance, utilised a commercial PE wax with different molecular weights, and in fact, they concluded that these differences in molecular weight are the main contributors to the segregation values. Not many other articles evaluated the effect of wax addition on storage stability were found in the literature, and among the few that did, they did not find a strong impact on the property [78].

The wax addition reduced the storage stability of the bitumen modified with PP. When the wax content is between 2 and 3% (PW2 and PW3), a slight reduction in the segregation value is noticed. Then, this reduction becomes more evident when the wax content is above 5%, reaching its minimum value at 7% (PW7), 26.22%, and bouncing back to 29.3% when 10% wax is added. The observed behaviour

makes it possible to infer that the pyrolytic wax can actually improve the storage stability of bitumen modified with plastic. This positive observation can be explained by the physical behaviour of the components during bitumen storage. When the PMB is stored at elevated temperatures, the polymer tends to float. This act causes the softening point of the top section to become harder, while the bottom section maintains the softening point of the neat bitumen. When the bitumen contains wax and polymer, although the top section still presents the floating polymer, at the bottom, the softening point has been increased too due to the wax. This response closes the gap between the top and bottom sections and contributes to the lowering of the segregation value.

A visual examination of the vertical sections of the samples used for the segregation test allows for the observation of the additives' behaviour during storage at high temperatures. Vertical sections of the samples C320, PP5, W7, and PW7 are depicted in Fig. 9. In the PP5 and PW7 samples, the floating polymer (red square) is vividly apparent. In contrast, homogeneity is observed in both unmodified and purely wax-processed bitumen, showing low segregation. In addition, these two samples lacked evidence of air pockets, unlike bitumen treated with PP and PP with HDPE wax (enclosed in blue in Fig. 9). During the beginning of the segregation test, the flocculation and floating of the plastic additives build a layer on top of the bitumen that prevents the escape of gases after heating. This may be the reason why PP5 and PW7 have void spaces between the bitumen and the floating polymer.

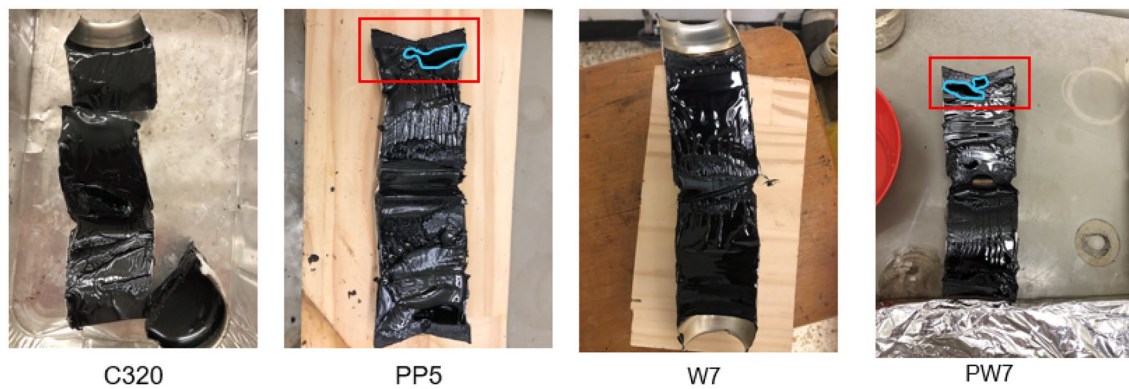


Fig. 9 Pictures after dividing the samples for segregation

Only the samples with the best performance on the previous tests will be submitted for additional testing on advanced bitumen properties. The PP5 sample was chosen for advanced tests on the binders that had only been modified with plastic. This sample had the lowest segregation when compared to the other plastic polymer-modified samples. The W7 sample was chosen for further testing of the wax-modified binders. The selection of W7 was mainly influenced by the fact that it allowed the largest plastic usage, allowing for greater recycling of plastic waste. Finally, among the samples treated with plastic waste and wax, the PW7 will be subjected to further advanced testing. This sample demonstrates the general benefits of polymers in bitumen: increased softening point and low penetration, as well as the lowest segregation when compared to other samples generated using a PP/wax combination.

3.3 Advanced Bitumen Tests

3.3.1 Fluorescent Microscope and FTIR

Images taken using a FM allow researchers to analyse the dispersion of additives in bitumen. This technique is widely utilised in the field since it aids in the visual differentiation

of polymer and bitumen in microscope pictures. In these circumstances, the plastic has a lighter luminosity, representing the polymer-rich phase, whereas the bitumen, representing the asphaltene-rich phase, has a darker glow. Figure 10 shows the FM of the neat bitumen, bitumen modified with 5% PP (PP5), modified with 7% wax (W7), and with a combined modification of 5% PP and 7% wax (PW7), and in Fig. 11, the images of the bottom and top sections of the segregation tests, for the same subsamples, are presented.

Figure 10 reveals that the wax distribution in the bitumen (W7) is homogeneous and that it has the same visual qualities as neat bitumen. Furthermore, no difference can be detected when comparing the bottom and top portions of the mentioned sample (Fig. 11), correlating with the segregation test results. When compared to the findings of Zhang et al. [34], the pyrolytic HDPE wax prepared in our work had the same segregation response as a commercial LDPE wax with a molecular weight of 4250. In the same study, it was discovered that the mentioned LDPE wax was capable of enhancing the service temperature of the bitumen at high temperatures.

The bitumen containing PP is easily identified in the FM images. Polymer component coalescence occurs, as does the swelling of individual plastic particles. This swelling effect

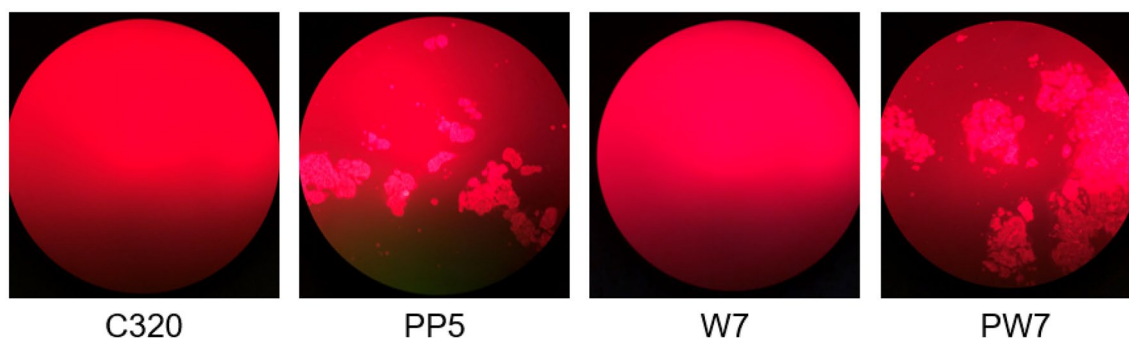


Fig. 10 Fluorescent microscope images (10 \times)

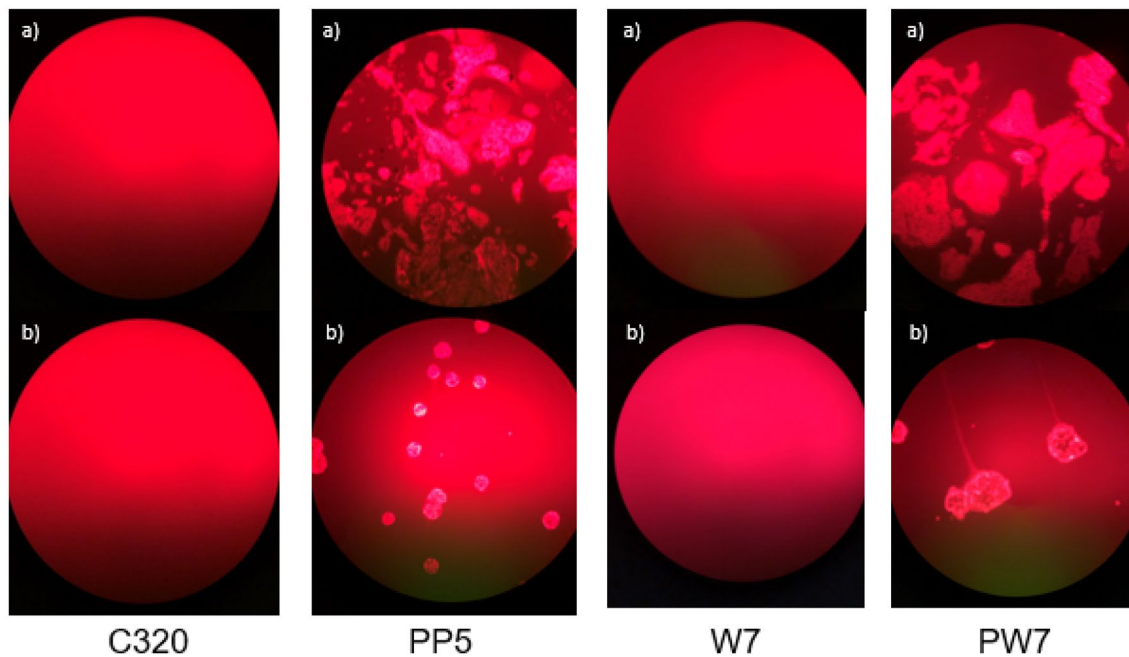


Fig. 11 Fluorescent microscope (10 \times) for the top (a) and bottom (b) sections obtained after the segregation test

is caused by the light maltene component of bitumen being absorbed by [79, 80]. According to Liang et al. [75], the coalescence process begins with plastic expansion and subsequent flocculation during the annealing stage. The result of these effects is the formation of a polymer network, which increases bitumen stiffness and improves softening point and penetration. When the segregation results in Fig. 11 are analysed, it is observed that the polymer-rich phase has migrated to the top portion, while the bottom segment retains a small proportion of polymer particles with small flocs. The latter observation is consistent with the fact that the dry polymer layer is located in the upper region of the segregation test, as indicated by the red squares in Fig. 9.

It is interesting to note that the addition of wax has increased the coalescence of the plastic particles. This can be seen by comparing the sizes of the plastic lumps in the FM images of samples PP5 and PW7, and it is more visible in Fig. 11 when comparing the bottom sections in the segregation samples. According to Rossi et al. [80], the wax may stimulate the polymer's absorption of certain bitumen components that were not absorbed when only the polymer was present. The aforementioned effect not only enlarges the swollen plastic particles, but it also promotes the formation of larger polymer-rich lumps. By increasing polymer coalescence in bitumen, the polymer network could be made more robust, which might enhance the binder properties.

The FTIR analysis shows that there is no considerable difference between the neat bitumen and the modified samples. Given that the amount of additive was low, it is not

surprising to observe that the formulated bitumen is chemically similar to the neat bitumen. Moreover, even the PW7, the bitumen with the largest amount of additives (5% PP and 7% pyrolytic HDPE wax), followed the same spectra profiles as the C320 bitumen. Although it is subtle, the only perceptible difference is around the spectrum at 1600 cm^{-1} (red circle), which corresponds to the aromatic portion C=C stretching. PP and pyrolytic HDPE wax do not present any peaks around the 1600 cm^{-1} range (Fig. 12 and Yuanita et al. [81]), and when they are added to the bitumen, they seem to cause a reduction in this corresponding absorbance. As previously stated, this difference is minimal, and thus the suggested modified bitumens are chemically appropriate for the formulation of asphalt mixes.

3.3.2 Viscosity and Mixing/Compaction Temperatures

Figure 13 depicts the viscosity of the samples at $135\text{ }^{\circ}\text{C}$ and $165\text{ }^{\circ}\text{C}$, demonstrating the polymer's clear detrimental effect. Although the addition of the polymer (PP5) resulted in a significant increase in viscosity, it remains less than the AASHTO M320 limit of 3 Pa.s. Even though the obtained number is consistent with earlier readings on softening point and penetration, it is not practical, as Köfteci et al. [82] contended, because it increases energy consumption and impairs workability. Previous literature reviews have highlighted this increase in viscosity as the primary adverse consequence of bitumen treated with plastic waste and its consequent influence on asphalt mixes [37, 38, 62]. Bitumen

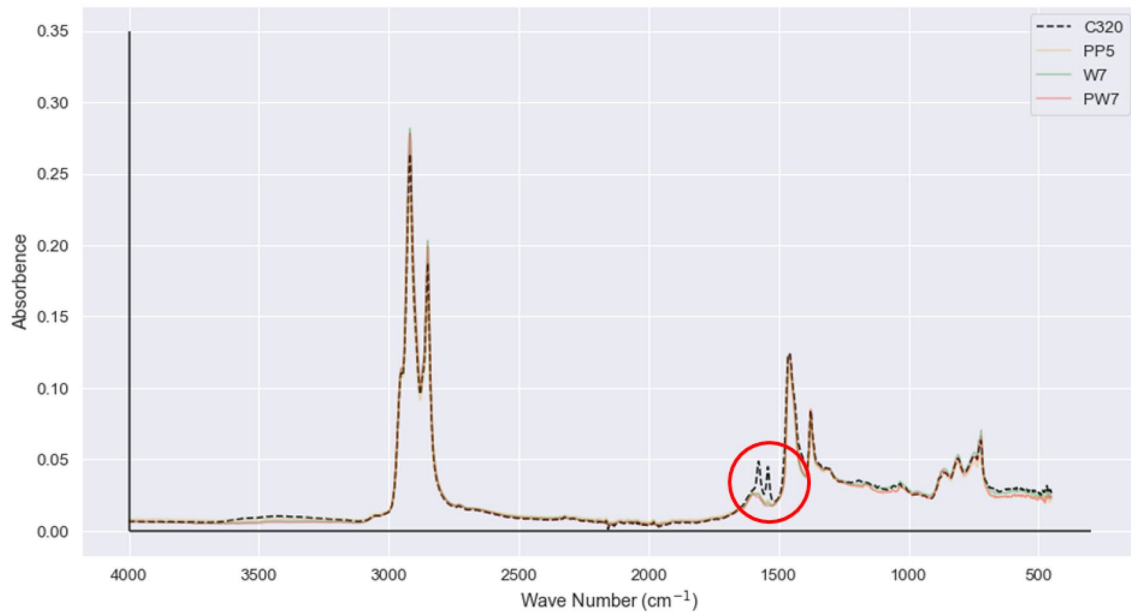
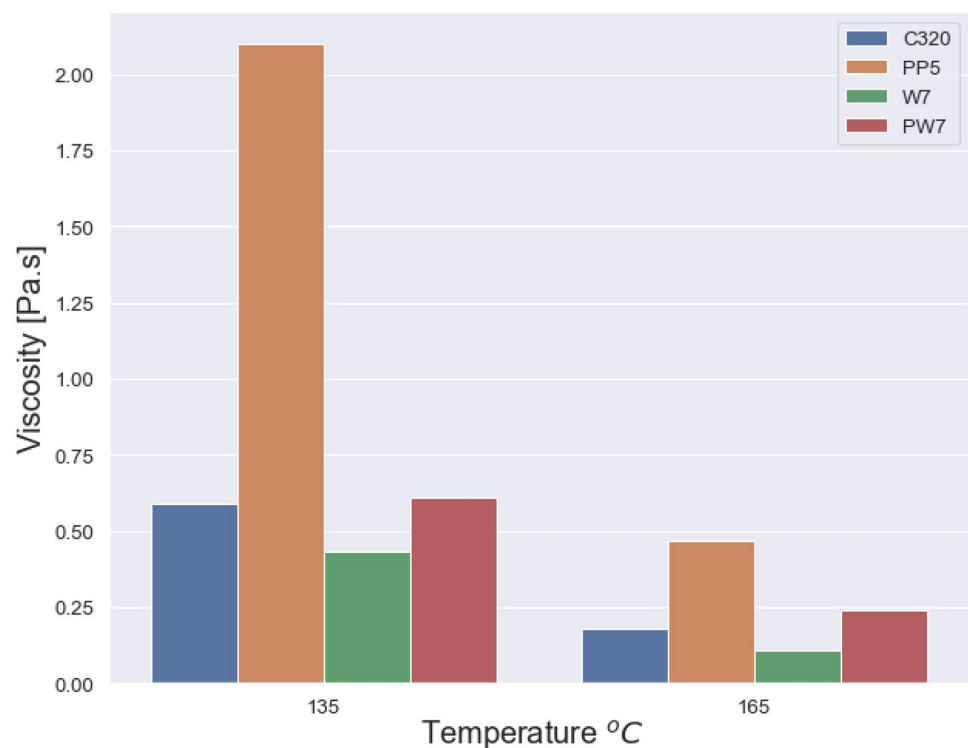


Fig. 12 FTIR for bitumen modified with polymer, wax, and polymer + wax

Fig. 13 Viscosity at 135 °C and 165 °C



with a high viscosity requires greater mixing and compaction temperatures, which also increases the bitumen content required for optimal asphalt mix and hence the overall cost of pavements [83].

When only the wax (W7) is added, it produces a binder with a lower viscosity than C320, which might also reduce the energy requirements for mixing and compacting asphalt

mixtures. This result is consistent with the findings of Shang et al. [20], who determined that pyrolytic wax generated from cross-linked polyethylene could reduce the binder's viscosity. Furthermore, it is consistent with other authors' remarks about the addition of commercial PE wax as a viscosity reducer [34, 84, 85]. According to Edwards et al. [72], the referred additives increase flow by softening the binder,

leading to a reduction in compaction and mixing temperatures. At the microstructure level, wax additives tend to feed the bitumen's maltene phase, resulting in a greater dispersion of asphaltene clusters and a change in the bitumen's flow and viscosity [86].

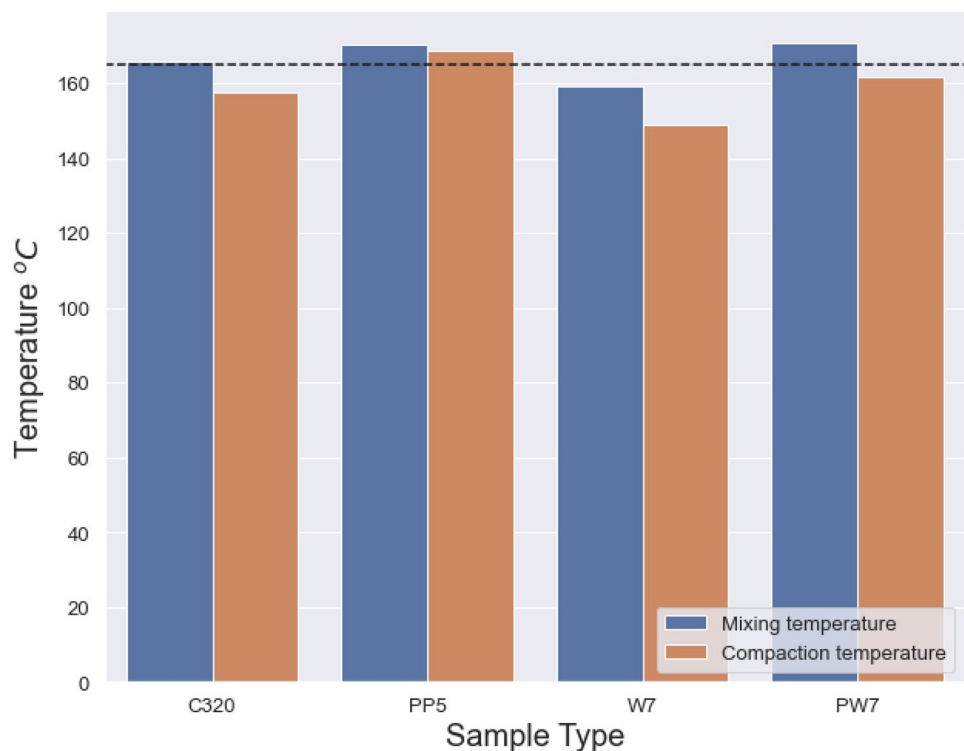
Surprisingly, the combination of polymer and wax (PW7) reduces the viscosity of the polymer-modified binder to a value that is somewhat higher than the viscosity of the unmodified binder. This observed behaviour is consistent with other studies that have investigated the effect of wax additives on polymer-modified bitumen with SBS [20, 78] and crumb rubber [87, 88]. At 165 °C, all samples met the Australian criteria for PMB containing elastomers and plastomers AGPT/T190 [58] and the Superpave standard AASHTO M332 [89], which require a maximum viscosity of 0.6 Pa.s. At this elevated temperature, the viscosity exhibits essentially the same behaviour as it did at 135 °C. The only difference is that the relative viscosity change between PW7 and C320 is greater at 165 °C than it is at 135 °C, and the gap between C320 and PP5 has shrunk.

Using the equiviscous method and the viscosity measurements recorded at 135 °C and 165 °C, it was possible to obtain the mixing and compaction temperatures (Fig. 14). C320 requires a mixing temperature of 165 °C and a compaction temperature of 157 °C degrees Celsius. When PP5 is introduced, mixing temperature increases by 5 °C, and compaction temperature increases by 11 °C. This increase is counterintuitive and may ultimately have

a detrimental impact on the net environmental impact of plastic waste incorporated into bitumen. In the life cycle analysis (LCA) performed by Yao et al. [90], although asphalt mixing and construction are the major contributors to the global warming potential, it seems that the viscosity increase produced by the addition of PET has not been taken into account. Nevertheless, this study still presents a positive net environmental impact due to the inclusion of the estimated service life of roads with plastic waste and avoided landfill emissions. Other similar studies have, likewise, overlooked the viscosity change and its effect on energy use, providing no definite conclusion about their environmental effects [91, 92].

The inclusion of wax (W7), as shown in Fig. 14, decreases the energy required for mixing and compaction. The mixing temperature is reduced by 6 °C compared with C320, while the compaction temperature is reduced by 8 °C. Although this reduction is not as significant as that of certain other commonly used commercial waxes [86], it is nonetheless remarkable given that the material is recycled plastic waste. Finally, while the addition of plastic and wax (PW7) does not result in a decrease in the temperature of the C320, it does result in a decrease in the energy required compared to the PMB (PP5). Compared to the latter, PW7 reduced the mixing and compaction temperatures by 1 °C and 7 °C, respectively. The positive impact of waxy additives on reducing energy demand has also been observed in the combination of PP and Fischer–Tropsch wax [93], SBS, and

Fig. 14 Mixing and compaction temperature



Sasobit-Advera-Rediset [94] and rubber with commercial warm-mix additives [95].

3.3.3 Stress Ratio

One of the most frequently cited problems with polymer-modified binders is that they reduce the binder's resistance low-temperature cracking [37]. Polymer-modified bitumen is typically stiffer than untreated bitumen, and at low temperatures, it becomes even stiffer. Because of this increased rigidity, a polymer-formulated binder is more susceptible to dynamic load-induced cracking. In Fig. 15, which depicts the strain–stress relationship of the formulated binders, this stiffness effect on bitumen modified with PP can be observed. In this figure, stiffness is represented by the slope of the plots during initial strains. PP5 was stiffer than C320, and the addition of wax increased this rigidity even more (PW7). When W7 and C320 are compared, although the stiffness rose when only wax was added to the bitumen (W7), the strain/stress curve remained close to C320's curve. This indicates that, while the stiffness of W7 has increased, it has not grown as aggressively as it did with PP5 and PW7, and that comparable cracking resistance might be expected with C320. The stress ratio result has also confirmed these discussed results (Table 4). C320 has the highest resistance to cold-temperature cracking, followed by W7. Moreover, despite having a similar value to W7, PP5 would be more susceptible to cracking at low temperatures due to its

Table 4 Stress ratio, consistency, and stiffness results

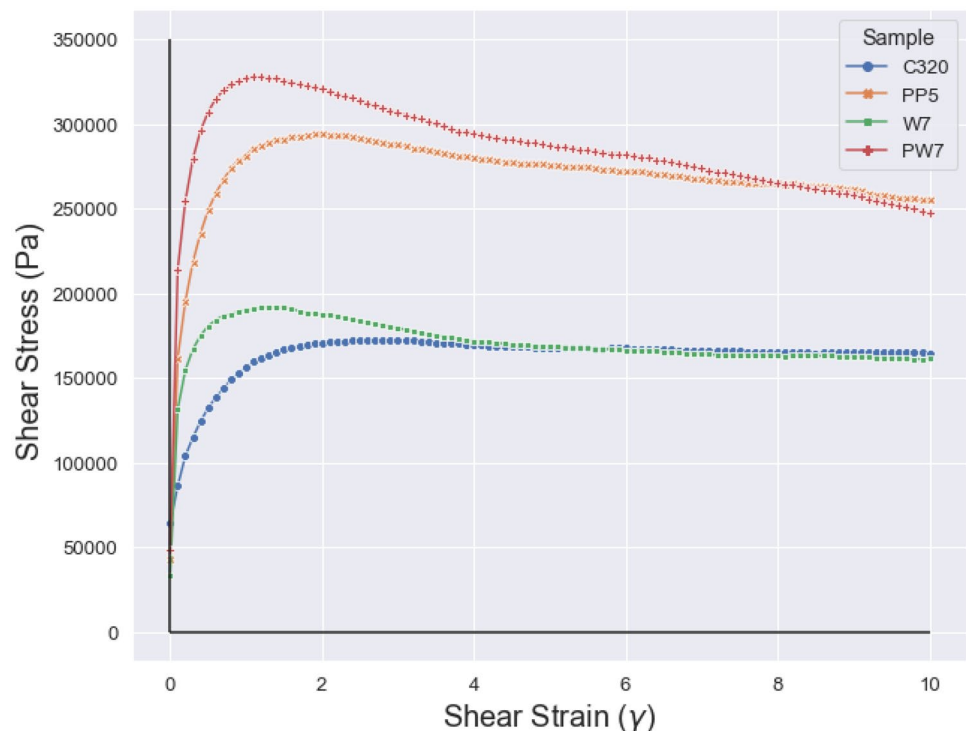
Sample type	Stress ratio at 10 °C	Consistency 6% at 60 °C (Pa.s)	Stiffness at 25 °C (kPa)
C320	0.96	504	42
PP5	0.89	1155	79
W7	0.9	1526	58
PW7	0.83	2698	100

stiffness. The same would happen with PW7, which had the lowest stress ratio (0.83).

3.3.4 Consistency at 6%

Overall, the results of the consistency test at 6% (Table 4) are consistent with the softening point observations. C320 had a low consistency value (504 Pa.s), which doubled when 5% PP was added (1155 Pa.s). Surprisingly, W7 has displayed greater consistency at 6% (1526 Pa.s) than PP5. This confirms that the addition of pyrolytic wax improves bitumen rutting resistance, and that this improvement is greater than that produced by unaltered plastic. It is also possible to infer that in rutting resistance, the crystallisation produced by the wax is more effective than the polymer-bitumen network produced by the PP. This observation is commendable because it indicates that the formulated wax presents the same benefits as plastic addition while eliminating or

Fig. 15 Strain/stress plot at 10 °C



reducing the most common drawbacks of polymer-modified binders: viscosity and storage stability. Last, as it was observed at the softening point, the consistency at 6% of PW7 seems to be the sum of the individual effects of wax and PP. This result is also noteworthy since it demonstrates that the wax can make polymer-modified bitumen 76% more resistant to rutting while reducing viscosity by 88% and segregation by 26%.

3.3.5 Stiffness

Stiffness is a general indicator of fatigue life. If a binder is overly stiff, it will not withstand dynamic loadings and will cause cracking in asphalt mixtures. The results of the stiffness test at 25 °C are displayed in Table 4. The addition of 5% PP increased bitumen stiffness by 88%, while the addition of 7% wax increased it by 38%. This is intriguing because, as is typical when additives are used in bitumen, a correlation is commonly observed between rutting resistance and stiffness. Bitumens with a higher rutting resistance are usually stiffer. Although W7 demonstrated greater rutting resistance than PP5, it also demonstrated lower stiffness. There is no clear explanation in the literature for why this could happen, so we hypothesise that the cause of this behaviour is the way the additive is spread and dissolved in the bitumen. The plastic additive forms a polymeric matrix that stiffens the bitumen when it is not completely dissolved in it. Wax, on the other hand, has been completely dissolved in the bitumen and, as a result of its crystallisation effect, has changed the microstructure of the binder. While both increase stiffness, the plastic's effect is stronger due to the macro matrix structure it forms. Wax, like the polymeric matrix, produces an internal structure characterised by its crystallisation effect, but its impact on final stiffness is not as strong because it allows for more flexibility. As was partly expected, the PW7 also presents an increase in stiffness, which can be characterised as the sum of the polymeric matrix and the wax crystallisation. Furthermore, because the wax promotes the formation of larger polymer flocs (as seen in the fluorescent microscope), it may cause a strengthening of the polymeric matrix that PP initially created.

4 Conclusion

The current work proposed a suitable bitumen additive through the pyrolysis of plastic. Although two different types of plastic were pyrolyzed (HDPE and PP), only the HDPE produced an acceptable wax with a low likelihood of lowering the bitumen's flashpoint. The effect of the formulated wax was evaluated on virgin bitumen C320 and polymer-modified bitumen with 5% PP. Softening point and consistency at 6% measurements demonstrated that adding wax

enhances the bitumen's resistance to high temperatures. In addition, although it increases the stiffness of the bitumen, it did not increase it as aggressively as the polymer. Thus, higher fatigue life and cold-temperature cracking resistance was better in the binder modified with wax than the PMB. Wax did not negatively affect segregation in C320 bitumen and reduced the PMB's segregation by 17% when the wax content was 7%. HDPE pyrolytic wax has also been shown to lower the viscosity of bitumen, which reduces the amount of energy needed to compact and mix asphalt mixtures. Based on these observations, the optimal wax concentration for virgin bitumen and PMB was determined to be 7%.

Pyrolytic wax was able to overcome two of the most significant drawbacks associated with the use of plastic waste as a bitumen additive: segregation and increased viscosity. Although lower amounts of wax do not have the same effect on the rigidity of the binder as ordinary PMBs, it still outperforms the PMB modified with 5%PP when the wax content is high (> 7%). These findings are significant in the current scientific context of plastic waste use in bitumen, and they may provide justification for using pyrolysis to increase the amount of plastic recycled in bituminous roadways.

Two of the most common criticisms of the wet mixing method are that the amount of plastic added is typically low—less than 5%—and that the wet technique requires more complex mixing equipment than the dry mixing method. Through this article, we've observed that pyrolysis pre-treatment could increase plastic usage and make it even greater than in the dry process. In the present investigation, for instance, the pyrolysis reaction was conducted at 350 °C for 30 min, which converted 90% of the plastic into wax. However, wax yields can be altered by adjusting the plastic's temperature and residence time in the pyrolysis reactor, so that it could increase or decrease the yields of gas, oil, and overall plastic utilisation. Utilising pyrolytic gas and oil can also improve the process's economics and mitigate its environmental impact. This is especially true considering the wax's intrinsic chemical composition did not change as a function of temperature or residence time. In addition, the use of wax does not increase the complexity of the binder formulation, as wax can be dissolved at 150 °C, the typical bitumen storage temperature, and high mixing speeds are not required.

In addition, it is interesting to observe how the wax can lower the temperature required for mixing and compaction. This characteristic has never been observed previously in bitumen mixtures with untreated plastics. Keeping this in mind, the given technique has the potential to reduce environmental effects by reducing plastic waste and consuming less energy during the asphaltic layer formulation process. However, this conclusion should be verified by an LCA study, which may be one of the future foci of this research. Additional research topics, including the study of HDPE

pyrolytic wax as a warm-mix additive, advanced rheology tests, and experiments on asphalt mixes, may be included in future studies to ensure the practicality of this material.

Annexes

Annex A One-Way ANOVA Penetration Measurement

	df	sum_sq	mean_sq	F	PR(> F)
Samples	14	11,271	805.068	1067.19	5.76E-126
Residual	129	97.3156	0.75438	NaN	NaN

Annex B Tukey Multiple Comparison of Means. Penetration Measurement

Group 1	Group 2	Mean dif-ference	p-adj	Lower	Upper	Reject
C320	PE5	- 25	0.001	- 26.416	- 23.584	True
C320	PE7	- 28.3222	0.001	- 29.7382	- 26.9062	True
C320	PP5	- 16.6556	0.001	- 18.0715	- 15.2396	True
C320	PP7	- 20.1333	0.001	- 21.3596	- 18.9071	True
C320	PW10	- 14.6333	0.001	- 16.0493	- 13.2174	True
C320	PW2	- 24.7222	0.001	- 26.1382	- 23.3062	True
C320	PW3	- 19.8222	0.001	- 21.2382	- 18.4062	True
C320	PW5	- 18.3889	0.001	- 19.8049	- 16.9729	True
C320	PW7	- 16.0556	0.001	- 17.4715	- 14.6396	True
C320	W2	- 9.2111	0.001	- 10.6271	- 7.7951	True
C320	W3	- 8.1	0.001	- 9.516	- 6.684	True
C320	W5	- 7.5778	0.001	- 8.9938	- 6.1618	True
C320	W7	- 7.2778	0.001	- 8.6938	- 5.8618	True
C320	Wax	- 35.7667	0.001	- 37.1826	- 34.3507	True
PE5	PE7	- 3.3222	0.001	- 4.7382	- 1.9062	True
PE5	PP5	8.3444	0.001	6.9285	9.7604	True
PE5	PP7	4.8667	0.001	3.6404	6.0929	True
PE5	PW10	10.3667	0.001	8.9507	11.7826	True
PE5	PW2	0.2778	0.9	- 1.1382	1.6938	False
PE5	PW3	5.1778	0.001	3.7618	6.5938	True
PE5	PW5	6.6111	0.001	5.1951	8.0271	True
PE5	PW7	8.9444	0.001	7.5285	10.3604	True
PE5	W2	15.7889	0.001	14.3729	17.2049	True
PE5	W3	16.9	0.001	15.484	18.316	True
PE5	W5	17.4222	0.001	16.0062	18.8382	True
PE5	W7	17.7222	0.001	16.3062	19.1382	True
PE5	Wax	- 10.7667	0.001	- 12.1826	- 9.3507	True

Group 1	Group 2	Mean dif-ference	p-adj	Lower	Upper	Reject
PE7	PP5	11.6667	0.001	10.2507	13.0826	True
PE7	PP7	8.1889	0.001	6.9626	9.4152	True
PE7	PW10	13.6889	0.001	12.2729	15.1049	True
PE7	PW2	3.6	0.001	2.184	5.016	True
PE7	PW3	8.5	0.001	7.084	9.916	True
PE7	PW5	9.9333	0.001	8.5174	11.3493	True
PE7	PW7	12.2667	0.001	10.8507	13.6826	True
PE7	W2	19.1111	0.001	17.6951	20.5271	True
PE7	W3	20.2222	0.001	18.8062	21.6382	True
PE7	W5	20.7444	0.001	19.3285	22.1604	True
PE7	W7	21.0444	0.001	19.6285	22.4604	True
PE7	Wax	- 7.4444	0.001	- 8.8604	- 6.0285	True
PP5	PP7	- 3.4778	0.001	- 4.7041	- 2.2515	True
PP5	PW10	2.0222	0.001	0.6062	3.4382	True
PP5	PW2	- 8.0667	0.001	- 9.4826	- 6.6507	True
PP5	PW3	- 3.1667	0.001	- 4.5826	- 1.7507	True
PP5	PW5	- 1.7333	0.0038	- 3.1493	- 0.3174	True
PP5	PW7	0.6	0.9	- 0.816	2.016	False
PP5	W2	7.4444	0.001	6.0285	8.8604	True
PP5	W3	8.5556	0.001	7.1396	9.9715	True
PP5	W5	9.0778	0.001	7.6618	10.4938	True
PP5	W7	9.3778	0.001	7.9618	10.7938	True
PP5	Wax	- 19.1111	0.001	- 20.5271	- 17.6951	True
PP7	PW10	5.5	0.001	4.2737	6.7263	True
PP7	PW2	- 4.5889	0.001	- 5.8152	- 3.3626	True
PP7	PW3	0.3111	0.9	- 0.9152	1.5374	False
PP7	PW5	1.7444	0.001	0.5182	2.9707	True
PP7	PW7	4.0778	0.001	2.8515	5.3041	True
PP7	W2	10.9222	0.001	9.6959	12.1485	True
PP7	W3	12.0333	0.001	10.8071	13.2596	True
PP7	W5	12.5556	0.001	11.3293	13.7818	True
PP7	W7	12.8556	0.001	11.6293	14.0818	True
PP7	Wax	- 15.6333	0.001	- 16.8596	- 14.4071	True
PW10	PW2	- 10.0889	0.001	- 11.5049	- 8.6729	True
PW10	PW3	- 5.1889	0.001	- 6.6049	- 3.7729	True
PW10	PW5	- 3.7556	0.001	- 5.1715	- 2.3396	True
PW10	PW7	- 1.4222	0.0478	- 2.8382	- 0.0062	True
PW10	W2	5.4222	0.001	4.0062	6.8382	True
PW10	W3	6.5333	0.001	5.1174	7.9493	True
PW10	W5	7.0556	0.001	5.6396	8.4715	True
PW10	W7	7.3556	0.001	5.9396	8.7715	True
PW10	Wax	- 21.1333	0.001	- 22.5493	- 19.7174	True
PW2	PW3	4.9	0.001	3.484	6.316	True
PW2	PW5	6.3333	0.001	4.9174	7.7493	True
PW2	PW7	8.6667	0.001	7.2507	10.0826	True
PW2	W2	15.5111	0.001	14.0951	16.9271	True
PW2	W3	16.6222	0.001	15.2062	18.0382	True
PW2	W5	17.1444	0.001	15.7285	18.5604	True
PW2	W7	17.4444	0.001	16.0285	18.8604	True
PW2	Wax	- 11.0444	0.001	- 12.4604	- 9.6285	True

Group 1	Group 2	Mean dif-ference	p-adj	Lower	Upper	Reject
PW3	PW5	1.4333	0.0442	0.0174	2.8493	True
PW3	PW7	3.7667	0.001	2.3507	5.1826	True
PW3	W2	10.6111	0.001	9.1951	12.0271	True
PW3	W3	11.7222	0.001	10.3062	13.1382	True
PW3	W5	12.2444	0.001	10.8285	13.6604	True
PW3	W7	12.5444	0.001	11.1285	13.9604	True
PW3	Wax	- 15.9444	0.001	- 17.3604	- 14.5285	True
PW5	PW7	2.3333	0.001	0.9174	3.7493	True
PW5	W2	9.1778	0.001	7.7618	10.5938	True
PW5	W3	10.2889	0.001	8.8729	11.7049	True
PW5	W5	10.8111	0.001	9.3951	12.2271	True
PW5	W7	11.1111	0.001	9.6951	12.5271	True
PW5	Wax	- 17.3778	0.001	- 18.7938	- 15.9618	True
PW7	W2	6.8444	0.001	5.4285	8.2604	True
PW7	W3	7.9556	0.001	6.5396	9.3715	True
PW7	W5	8.4778	0.001	7.0618	9.8938	True
PW7	W7	8.7778	0.001	7.3618	10.1938	True
PW7	Wax	- 19.7111	0.001	- 21.1271	- 18.2951	True
W2	W3	1.1111	0.3093	- 0.3049	2.5271	False
W2	W5	1.6333	0.0091	0.2174	3.0493	True
W2	W7	1.9333	0.001	0.5174	3.3493	True
W2	Wax	- 26.5556	0.001	- 27.9715	- 25.1396	True
W3	W5	0.5222	0.9	- 0.8938	1.9382	False
W3	W7	0.8222	0.7615	- 0.5938	2.2382	False
W3	Wax	- 27.6667	0.001	- 29.0826	- 26.2507	True
W5	W7	0.3	0.9	- 1.116	1.716	False
W5	Wax	- 28.1889	0.001	- 29.6049	- 26.7729	True
W7	Wax	- 28.4889	0.001	- 29.9049	- 27.0729	True

Annex C One-Way ANOVA Softening Point Measurement

	df	sum_sq	mean_sq	F	PR(>F)
Samples	13	5074.1022	390.31556	1479.8694	1.05E-85
Residual	76	20.045	0.26375	NaN	NaN

Annex D Tukey Multiple Comparison of Means. Softening Point Measurement

Group 1	Group 2	Mean dif-ference	p-adj	Lower	Upper	Reject
C320	PE5	21	0	19.9727	22.0273	True
C320	PE7	25.3167	0	24.2894	26.344	True
C320	PP5	5.05	0	4.0227	6.0773	True

Group 1	Group 2	Mean dif-ference	p-adj	Lower	Upper	Reject
C320	PP7	18.45	0	17.5603	19.3397	True
C320	PW10	11.85	0	10.8227	12.8773	True
C320	PW2	4.3667	0	3.3394	5.394	True
C320	PW3	5.0333	0	4.006	6.0606	True
C320	PW5	7.3667	0	6.3394	8.394	True
C320	PW7	7.7667	0	6.7394	8.794	True
C320	W2	1.2333	0.0059	0.206	2.2606	True
C320	W3	2.9833	0	1.956	4.0106	True
C320	W5	5.25	0	4.2227	6.2773	True
C320	W7	7.3	0	6.2727	8.3273	True
PE5	PE7	4.3167	0	3.2894	5.344	True
PE5	PP5	- 15.95	0	- 16.9773	- 14.9227	True
PE5	PP7	- 2.55	0	- 3.4397	- 1.6603	True
PE5	PW10	- 9.15	0	- 10.1773	- 8.1227	True
PE5	PW2	- 16.6333	0	- 17.6606	- 15.606	True
PE5	PW3	- 15.9667	0	- 16.994	- 14.9394	True
PE5	PW5	- 13.6333	0	- 14.6606	- 12.606	True
PE5	PW7	- 13.2333	0	- 14.2606	- 12.206	True
PE5	W2	- 19.7667	0	- 20.794	- 18.7394	True
PE5	W3	- 18.0167	0	- 19.044	- 16.9894	True
PE5	W5	- 15.75	0	- 16.7773	- 14.7227	True
PE5	W7	- 13.7	0	- 14.7273	- 12.6727	True
PE7	PP5	- 20.2667	0	- 21.294	- 19.2394	True
PE7	PP7	- 6.8667	0	- 7.7563	- 5.977	True
PE7	PW10	- 13.4667	0	- 14.494	- 12.4394	True
PE7	PW2	- 20.95	0	- 21.9773	- 19.9227	True
PE7	PW3	- 20.2833	0	- 21.3106	- 19.256	True
PE7	PW5	- 17.95	0	- 18.9773	- 16.9227	True
PE7	PW7	- 17.55	0	- 18.5773	- 16.5227	True
PE7	W2	- 24.0833	0	- 25.1106	- 23.056	True
PE7	W3	- 22.3333	0	- 23.3606	- 21.306	True
PE7	W5	- 20.0667	0	- 21.094	- 19.0394	True
PE7	W7	- 18.0167	0	- 19.044	- 16.9894	True
PP5	PP7	13.4	0	12.5103	14.2897	True
PP5	PW10	6.8	0	5.7727	7.8273	True
PP5	PW2	- 0.6833	0.5569	- 1.7106	0.344	False
PP5	PW3	- 0.0167	1	- 1.044	1.0106	False
PP5	PW5	2.3167	0	1.2894	3.344	True
PP5	PW7	2.7167	0	1.6894	3.744	True
PP5	W2	- 3.8167	0	- 4.844	- 2.7894	True
PP5	W3	- 2.0667	0	- 3.094	- 1.0394	True
PP5	W5	0.2	1	- 0.8273	1.2273	False
PP5	W7	2.25	0	1.2227	3.2773	True
PP7	PW10	- 6.6	0	- 7.4897	- 5.7103	True
PP7	PW2	- 14.0833	0	- 14.973	- 13.1937	True
PP7	PW3	- 13.4167	0	- 14.3063	- 12.527	True
PP7	PW5	- 11.0833	0	- 11.973	- 10.1937	True
PP7	PW7	- 10.6833	0	- 11.573	- 9.7937	True
PP7	W2	- 17.2167	0	- 18.1063	- 16.327	True

Group 1	Group 2	Mean difference	p-adj	Lower	Upper	Reject
PP7	W3	-15.4667	0	-16.3563	-14.5777	True
PP7	W5	-13.2	0	-14.0897	-12.3103	True
PP7	W7	-11.15	0	-12.0397	-10.2603	True
PW10	PW2	-7.4833	0	-8.5106	-6.456	True
PW10	PW3	-6.8167	0	-7.844	-5.7894	True
PW10	PW5	-4.4833	0	-5.5106	-3.456	True
PW10	PW7	-4.0833	0	-5.1106	-3.056	True
PW10	W2	-10.6167	0	-11.644	-9.5894	True
PW10	W3	-8.8667	0	-9.894	-7.8394	True
PW10	W5	-6.6	0	-7.6273	-5.5727	True
PW10	W7	-4.55	0	-5.5773	-3.5227	True
PW2	PW3	0.6667	0.5966	-0.3606	1.694	False
PW2	PW5	3	0	1.9727	4.0273	True
PW2	PW7	3.4	0	2.3727	4.4273	True
PW2	W2	-3.1333	0	-4.1606	-2.106	True
PW2	W3	-1.3833	0.001	-2.4106	-0.356	True
PW2	W5	0.8833	0.169	-0.144	1.9106	False
PW2	W7	2.9333	0	1.906	3.9606	True
PW3	PW5	2.3333	0	1.306	3.3606	True
PW3	PW7	2.7333	0	1.706	3.7606	True
PW3	W2	-3.8	0	-4.8273	-2.7727	True
PW3	W3	-2.05	0	-3.0773	-1.0227	True
PW3	W5	0.2167	1	-0.8106	1.244	False
PW3	W7	2.2667	0	1.2394	3.294	True
PW5	PW7	0.4	0.9848	-0.6273	1.4273	False
PW5	W2	-6.1333	0	-7.1606	-5.106	True
PW5	W3	-4.3833	0	-5.4106	-3.356	True
PW5	W5	-2.1167	0	-3.144	-1.0894	True
PW5	W7	-0.0667	1	-1.094	0.9606	False
PW7	W2	-6.5333	0	-7.5606	-5.506	True
PW7	W3	-4.7833	0	-5.8106	-3.756	True
PW7	W5	-2.5167	0	-3.544	-1.4894	True
PW7	W7	-0.4667	0.9475	-1.494	0.5606	False
W2	W3	1.75	0	0.7227	2.7773	True
W2	W5	4.0167	0	2.9894	5.044	True
W2	W7	6.0667	0	5.0394	7.094	True
W3	W5	2.2667	0	1.2394	3.294	True
W3	W7	4.3167	0	3.2894	5.344	True
W5	W7	2.05	0	1.0227	3.0773	True

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Data availability The authors hereby affirm that the data supporting the results of this study are accessible within the article and/or accompanying supplementary materials.

Declarations

Conflict of Interest Not applicable.

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