Deriving and Characterising Alternative Bitumen from Waste Plastics

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ABSTRACT

This study presents research on laboratory production and experimental characterisation of an alternative bitumen using municipal waste plastics. Six different waste plastics (A1 - A6) produced by a local waste recycling manufacturers were selected and characterised to investigate their feasibility in modifying the bitumen binders. Thermal characteristics were firstly obtained using Differential Scanning Calorimetry (DSC) device and the chemical functional groups were identified by Fourier Transform Infrared Spectroscopy (FT-IR) test to determine the plastic types existing in the recycled plastics. Then the rheological properties of the bitumen modified with two nominated plastic waste (A1 and A2) were examined using the Dynamic Shear Rheometer (DSR) device by conducting frequency sweep tests. Additionally, the engineering performance of waste plastics-derived bitumen was also obtained and compared against the control bitumen, including fatigue, rutting and healing performance using Time Sweep (TS) test, Multiple Stress Creep and Recovery (MSCR) test and Healing test, respectively.

Results show that A1and A2 consist of low-density polyethene (LDPE) and polypropylene (PP), respectively. The recycled waste plastic A5 and A6 (both classified under the same category but collected from different plants and batches) are mainly consisting of LDPE. Whereas, other recycled plastics (A3 and A4) consist of a variety of materials and impurities. Thus, A1 and A2 were chosen as bitumen binder extenders. A1-modified bitumen exhibited more elastic and less viscous behaviour than the control bitumen, showed by increased shear modulus and reduced phase angle. Whereas, A2 (consisting of PP) caused a significant drop in the shear modulus. Both recycled LDPE and PP-modified bitumen had a substantially improved resistance to rutting and fatigue cracking compared to the control bitumen. Furthermore, waste LDPE-modified bitumen sustained increased healing potential compared to waste PP-modified bitumen, where the latter did not show noticeable improvement to the healing performance.

1. INTRODUCTION

Plastics are multi-purpose products used in all aspects of urban life, from food containers to construction materials and automobiles industries. Therefore, they are produced in large quantities to cover these high demands. Plastic groups in total non-fibre plastics production are classified from largest to smallest quantities into polyethene (PE) (36%), polypropylene (PP) (21%), and polyvinyl chloride (PVC) (12%), followed by polyethene terephthalate (PET), polyurethane (PU), and polystyrene (PS) (<10% each). About 2.8 Mt of waste plastics is generated per annum in the UK alone, making 7.3% of the total demand for plastic in Europe. However, the end-of-use for these materials is a cause for environmental concerns. The plastic is a non-biodegradable product that negatively impacts the environment if disposed of improperly, such as disposal in waste land-fills. It is estimated that eight million tons of plastic waste are dumped into the ocean annually around the globe. However, since the plastic is a very versatile material, it is not possible to cut the use of plastics completely. The solution to ending the plastic problem does not involve the reduced use alone, but also, finding better ways to recycle waste plastics in bulk quantities [1-6]. Recycling of waste plastics can delay their end of useful lifetime and discarding process. The use of plastics in construction applications has the longest useful lifespan, in which the materials take decades before they need to be recycled/disposed of again. This is due to the long lifespan of construction products, whereas, other products of high plastics demand, such as packaging, mostly have less than one year of product life. Thus, these plastics will need to be processed/discarded again in a short period, causing an economic and environmental burden [5].

The use of plastics in the manufacturing of flexible pavements helps to consume large quantities of waste plastics and reduces demands for producing petroleum-based bitumen. Therefore, it solves an environmental problem and helps in saving natural resources. Several studies investigated the utilisation of waste plastics in flexible pavement mixtures. The incorporation of plastics in asphalt mixtures was introduced in three forms: (1) partial replacement of aggregate with recycled plastic shreds of similar particle size [7]; (2) partial replacement of bitumen binder by recycled plastic shreds [8]; (3) waste plastic is introduced into the binder in the powdered form [9]. The percentage of plastics replacing the aggregate was found to reach up to 30% by aggregate weight. However, some asphalt mixtures require increasing the binder content when replacing the traditional aggregate with plastics waste. On the other hand, the percentage of plastic to replace the bitumen was found to reach up to 10% by bitumen weight [6, 8]. Most studies reported reduced permanent deformations (in the form of reduced rutting and low-temperature cracking of pavement surface) upon adding recycled waste plastics [8]. Literature search marked the use of several types of

plastics in asphalt mixtures, including Low- and High-Density Polyethylene (LDPE and HDPE, PET [10, 11], PVC, and PP [8]. LDPE and HDPE possess a relatively low softening point temperature of 130 °C, and thermal degradation temperature of more than 180 °C. Therefore, if used to replace aggregate in asphalt mixtures, PE is compatible with the current mixing procedures and does not require any modification to existing asphalt production plant facilities and techniques [4, 7]. Whereas, PET-aggregate modified mixtures exhibited higher fatigue lives compared to mixtures without PET [12]. Although these studies examined the potential application of first-use plastics in pavement manufacturing, incorporating the *recycled* waste of these plastics is still limited.

The use of waste plastics as bitumen modifier was hindered due to several knowledge gaps that need to be addressed. Uncertainties related to waste plastics use include (1) effect of chemical and physical characteristics of plastics on the bitumen binders upon mixing (e.g., the impact of particle size of plastic shreds on mixing time to achieve full dispersion and to avoid phase-separation between asphalt binders and plastic); (2) the suitability of current mixing procedures and equipment for handling plastic-modified binders and asphalt mixtures is unknown (e.g., the mixing temperature and time required for full melting and dispersion of plastic into bitumen, and time needed for laying-out and compaction in the field before the mixture hardens); (3) availability of recycled waste plastics in large bulk quantities with maintaining consistent properties for different patches and produce; (4) apart from technical factors, concerns over the use of plastics in asphalt pavement also manifest from their potential of causing environmental problems, such as run-off pollutants and leaching [4]. In addition, the focus of existing studies is centred on the mechanical characterisation and performance of mixtures upon the introduction of waste plastics with few studies addressing the effects of plastics at binder level [13, 14]. Therefore, fundamental research on the bitumen interaction with waste plastics is still needed.

The current study aims to investigate the feasibility of modifying bitumen with waste plastics. This is done by examining the chemical and physical characteristics of waste plastics produced by local waste recycling suppliers and study their effect on the bitumen binder upon mixing at high temperatures. The physical properties, such as the thermal behaviour of waste plastics, are examined using the Differential Scanning Calorimetry (DSC) device to select a convenient mixing temperature. The Fourier Transform Infrared Spectroscopy (FT-IR) test is employed to identify the chemical functional groups, and accordingly, determine the exact plastic types within the produced batches of recycled waste plastics. After that, the rheological properties of bitumen modified with waste plastic are examined, where The Dynamic Shear Rheometer (DSR) device is employed to run frequency sweep tests. Then, the engineering performance of waste plastics-modified bitumen is assessed. Fatigue resistance, rutting performance and healing potential of the plastic-modified binders are considered in this category. This is done by conducting Time Sweep (TS) tests; Multiple Stress Creep and Recovery (MSCR) tests and Healing tests.

This study focuses on the thermal and chemical characterisation of several locally-produced recycled waste plastics for asphalt pavements. The study compares the preliminary mechanical performance of two waste plastic-modified bitumen at the unaged condition. Therefore, a follow-up study will be needed to examine the field performance of waste plastics-modified bitumen after-mixing and layout and the long-term field performance.

2. MATERIALS

2.1 Recycled plastic

Six types of recycled plastics are evaluated in this study: A1, A2, A3, A4, A5, and A6 (shown in Figure 1). The first type, A1 consists of thin white or brown pellets, A2 consists of thick green pellets, A3 consists of white or clear flakes, A4 is mixed-coloured particles of plastic shreds, and A5 and A6 both consists of mixed-coloured plastic particles, but the latter is smaller in size than A4. All the plastics types are supplied from local recycled-plastic providers. The recycled-plastic samples were not produced for a specific application; therefore, they are not provided with technical data sheets to indicate their chemical composition, origin, or physical properties.
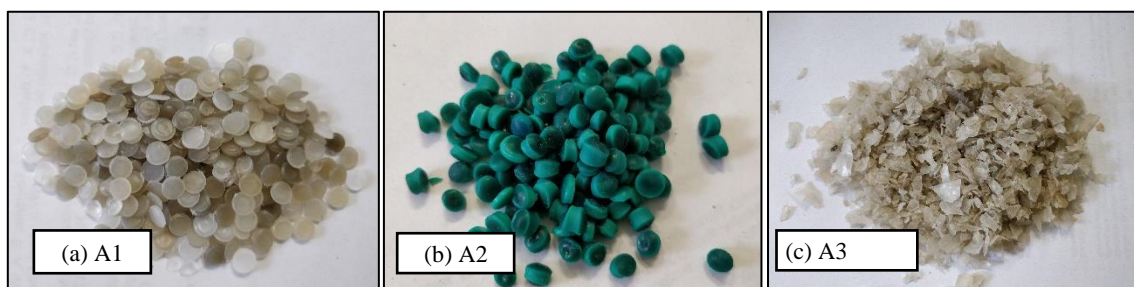

(a) A1   (b) A2   (c) A3

Figure 1: Recycled plastic types as provided by the supplier

2.2 Bitumen binder

The prime asphalt binder used in this study is known as Bitumen X70 classified as a 40/100 according to BSEN14023:2010 for general asphalt applications and road constructions. It is used as a base (control) binder to examine the effect of adding the recycled plastics. Table 1 lists the engineering specifications of the control binder, as mentioned in the technical sheet.

Table 1. Conventional properties of the control binder

Property	Value
Penetration @ 25 °C (0.1 mm)	45–80
Softening point (°C)	≥ 45
Flash point (°C)	> 250
Force Ductility @ 5 °C (J/cm2)	> 3

2.3 Waste plastic modified-bitumen

After evaluating their thermal characteristics and chemical composition by FT-IR, two plastic waste types are selected to be mixed with the control binder (A1 and A2), to evaluate their engineering performance (further details about the selection criteria are illustrated in the following sections). To develop the waste plastic-modified bitumen, control bitumen X70 is mixed with the plastic waste by mass percentages. The optimum mixing percentage of the plastic waste is selected by mixing different plastic percentages (5, 7, 10 and 12 % by w/w of X70 binder) with the control binder and testing them by frequency sweep tests at various temperatures. It is found that adding 6 % of recycled plastic achieved higher complex shear modulus with considerably good phase angle. Therefore, X70 bitumen is mixed with 6 % plastic waste using the mechanical high-shear rotational mixer at high mixing temperature. Mixing of the binder with plastic waste at high-temperature conditions is vital to allow the binder compounds to be incorporated uniformly with the plastic and to sustain a homogenous mixture. There are several mixing schemes suggested by existing studies for plastic, bio-oil and polymer components [4, 7, 8, 15-20]. Accordingly, to ensure a complete melting and mixing of plastic-waste with bitumen, a temperature of 180 °C and mixing speed of 900 rpm are maintained for 60 minutes to avoid excessive ageing of the samples. Finally, specimens are poured into DSC moulds of 8- and 25-mm diameters.

3 EXPERIMENTAL METHODS
3.1 Thermal characterisation of waste plastics by Differential Scanning Calorimetry (DSC)

Recycled plastic samples are tested using the DSC device to observe their thermal stability and phase changes upon heating. The recycled plastics are obtained from a local waste plastic supplier, and the chemical composition of these recycled materials are unknown. The DSC test helps identify types of plastics present in these samples by subjecting them to a dynamic thermal condition in which temperature increases at a constant rate, then any heat flow is recorded and attributed to the thermal phases of tested specimen. When used under the dynamic thermal condition, the DSC technique can measure the melting temperature and crystallinity of polymers.

The DSC measurements are performed using METTLER TOLEDO gas control CG10 - heat-flux DSC, aluminium crucibles (without lid), and a purge gas of pure nitrogen, at a flow rate of 10 °C/min. The device uses a reference empty aluminium crucible, which sits on the sensors to monitor the differential heat flow between the reference crucible and the crucible carrying the sample. Specimens masses are kept in the range of 5.1 to 5.8 mg.

DSC specimens containing waste plastics are cooled from room temperature (25 °C) to -50 °C, to start the test. Some samples are heated to a temperature of 190 °C, then cooled down to -50 °C before beginning DSC test to remove thermal history effects. In other words, all the samples will have a similar thermal history, and there will be no difference in thermal behaviour due to past thermal events. Once the start temperature (-50 °C) is set, samples are heated up to 300 °C to pass their melting point and potential degradation point.

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Waste plastic samples are tested twice to observe the repeatability of results. The replicates for each plastic type found to be identical in their thermal behaviour, which indicates that the results are reliable. The samples are named according to their visual appearances and colours. The visual appearances of samples tested by DSC are listed in Table 2. A4, A5 and A6 appear to consist of different components according to their visual appearance; therefore, random pieces are collected and tested separately. Moreover, representative samples consisting of various elements (collected from samples of multi colours) are also included in the DSC testing program.

Table 2. Visual description and mass of waste plastic samples tested by DSC device

Sample	Description	Mass (mg)
A1-01-CLR	White/colourless A1 sample	5.3
A1-02-CLR	White/colourless A2 sample	5.3
A1-01-DRK	Dark/brown A1 sample	5.3
A1-02-DRK	Dark/brown A1 sample	5.3
A2-01	Green, A2 sample	5.2
A2-02	Green, A2 sample	5.3
A3-01	Colourless A3 sample in flakes	5.4
A3-02	Colourless A3 sample in flakes	5.3
A4-01	A random mixture of A4 samples (mixed colour)	5.8
A4-02	A random mixture of A4 samples (mixed colour)	5.6
A4-BK	Black, a sample chosen randomly from A4.	5.7
A4-CLR	Colourless, a sample chosen randomly from A4.	5.5
A4-Y	Yellow, a sample chosen randomly from A4.	5.5
A5	A random mixture of A5	5.7
A6-Y	Yellow, a sample chosen randomly from A6.	5.3
A6-W	White, a sample chosen randomly from A6.	5.4
A6-BK	Black, a sample chosen randomly from A6.	5.1

3.2 Chemical characterisation of waste plastics by Fourier Transform Infrared Spectroscopy (FT-IR) test

FT-IR test is utilised to determine the chemical composition and functional groups present in the recycled waste plastics. FT-IR test results for waste plastic specimens will help identify the plastic-type used in their manufacturing by comparing the functional groups obtained with the signature (standard) functional groups of polymers in a fast, efficient, and non-destructive procedure [20]. The chemical structures of HDPE and LDPE are challenging to differentiate. However, recent research studies distinguished between the two polymers using FT-IR spectra [21].

The FT-IR measurements are conducted using PerkinElmer FT-IR Spectrometer device. The device is set to scan in a range of (400-4000) cm^{-1}, with a scanning frequency number of 16 and a resolution of 4 cm^{-1}. Background scan is set before testing the samples. Names and description of visual appearance for the samples scanned under the FT-IR device are listed in Table 3.

Table 3. Identification and visual description of waste plastics tested using FT-IR device

Sample	Description
A1-CLR	White/colourless A1 sample
A1-DRK	Dark/brown A1 sample
A2	Green, A2 sample
A3	A sample is chosen randomly from A3
A4-B	Blue, a sample chosen randomly from A4.
A4-BK	Black, a sample chosen randomly from A4.
A4-CLR	Blue, a sample chosen randomly from A4.
A4-G	Colourless, a sample chosen randomly from A4.
A4-GY	Grey, a sample chosen randomly from A4.
A4-R	Red, a sample chosen randomly from A4.
A4-W	White, a sample chosen randomly from A4.
A4-Y	Yellow, a sample chosen randomly from A4.
A5-B	Blue, a sample chosen randomly from A5.
A5-CLR	Colourless, a sample chosen randomly from A5.
A5-G	Green, a sample chosen randomly from A5.
A5-R	Red, a sample chosen randomly from A5.
A5-Y	Yellow, a sample chosen randomly from A5.
A6-BK-01	Black, a sample chosen randomly from A6.
A6 -BK-02	Black, a sample chosen randomly from A6.
A6-B	Blue, a sample chosen randomly from A6.
A6-CLR	Colourless, a sample chosen randomly from A6.

A6-G-01	Green, a sample chosen randomly from A6.
A6-R	Red, a sample chosen randomly from A6.
A6-W	White, a sample chosen randomly from A6.

3.3 Rheological characterisation of waste plastics-modified bitumen: Frequency sweep test by Dynamic Shear Rheometer (DSR)

Inclusion of waste plastics into bitumen can affect the rheological properties of resulting bitumen, leading to a different viscoelastic performance when compared to the control binder. The frequency sweep test is used to characterise the rheological behaviour of waste plastics-modified bitumen by examining the shear modulus and phase angle at undamaged conditions. Frequency sweep tests are conducted at a temperature range of 10 – 70 °C with temperature intervals of 10 °C. The selected temperature range is chosen to simulate the wide temperatures range in the field. The strain level at temperatures from 10-30 °C is specified to be 0.3% strain to induce a low torque, because bitumen binders become stiffer at low temperatures, so a low strain is applied to prevent cracking. Whereas, for temperatures exceeding 30 °C, 0.5% strain is applied as the bitumen binders are softer (less viscous), which implies a higher tolerance to strain, so the binders will not fail by cracking [22]. The frequencies set to be 0.1 – 25 Hz. Undamaged shear moduli and phase angles were obtained from master curves of temperatures using the time-temperature superposition principle (TTSP) [23, 24].

3.4 Engineering performance characterisation of waste plastics-modified bitumen

Three testing schemes are employed to evaluate the engineering performance of waste plastics-modified bitumen using DSR device: (1) time sweep (TS) test, to examine the fatigue cracking resistance of the modified bitumen binders under strain-controlled cyclic loading conditions; (2) multiple stress creep and recovery (MSCR) test to determine the rutting resistance performance; and (3) healing test to determine the percentage of recovery and healing capability of waste plastics-modified bitumen upon unloading.

The TS test is performed at a strain level of 5%, at an intermediate temperature of 20 °C and a frequency of 10 Hz, up to 24,000 loading cycles. The previous study by the authors conducted on the same source of bitumen indicates that it tends to experience fatigue cracking at a loading cycle number of 24,000 [25]. Therefore, the same loading cycle (N= 24,000) is selected as TS loading limit in this testing scheme. The shear moduli and phase angles are recorded throughout the test. The fatigue cracking is quantified by using DSR-C growth model based on the dissipated strain energy equilibrium principle and damage mechanics [25]. The study employed the DSR to determine the shear moduli and phase angles at both undamaged and damaged conditions. Then, the fatigue damage is quantified directly by the predicted crack length (c_p), illustrated in Eq. (1).

$$c_p = \left[1 - \left(\frac{|G_N^*|/\sin(\delta_N)}{|G_o^*|/\sin(\delta_o)} \right)^{\frac{1}{4}} \right] r_o \tag{1}$$

where,

r_o = original radius of the sample (4 mm)

$|G_N^*|$, $|G_o^*|$ = shear moduli at damaged and undamaged conditions, respectively

δ_N, δ_o = phase angles of damaged and undamaged conditions, respectively.

c_p = predicted crack length at the Nth loading cycle of the time sweep fatigue test.

Binders modified with plastic can be prone to rutting at high temperatures. Hence, it is imperative to evaluate the extent of damage that can occur [26]. The MSCR test is utilised to characterise the rutting resistance performance of plastic-modified binders effectively. The test method follows the standard test specification of ASTM standard D7405-15 [27]. MSCR test records two essential parameters: percentage recovery (% R) and non-recovery compliance (Jnr) of asphalt binders.

Healing tests are carried out to examine how asphalt binders can recover their mechanical properties (e.g., stiffness and strength) under loading in a fatigue test [28, 29]. The ability to self-heal is controlled by many factors (i.e. viscosity level, temperature and rest duration) and has been associated with improving the longevity of a roads service life. Therefore, an investigation into this field is important to identify which binders can self-heal to reduce the need to carry out replacement or maintenance of roads. The self-healing capability of waste plastic-derived bitumen will be examined using DSR healing test. The test consists of two loading stages of strain-controlled time sweep tests separated by a rest period (unloading stage) where there is no torque/strain applied for 20 minutes. The time sweep stages are conducted with a frequency of 20 Hz and 5 % strain at an intermediate temperature of 20 °C. The healing

is quantified by measuring the shear moduli before and after the healing stage, and the Healing Index (*HI*) is illustrated in Eq. 2.

$$HI = \frac{G^*_{after} - G^*_{before}}{G^*_{before}} * 100 \qquad (2)$$

where G^*_{after} is the complex shear modulus of the self-healed binder, measured in MPa, and G^*_{before} is the complex shear modulus before healing, measured in MPa.

4   THERMAL AND CHEMICAL CHARACTERISATION OF WASTE PLASTICS

Recycled waste plastics are recovered from municipal land-fills that commonly come from various sources and contain several components, such as different plastic types and impurities (wood, rocks, etc.). Therefore, to utilise these waste-plastics in the flexible pavement industry, these materials should be identified to characterised their chemical composition and physical properties. The thermal behaviour (melting point, degradation point and thermal stability) of waste plastics is examined using the DSC. The chemical composition (represented by the chemical functional groups) is identified by conducting FT-IR scanning. Results for each plastic-type are detained in the following sub-sections.

4.1 DSC and FT-IR results for plastic-type A1

Figure 2 shows the DSC test results for two samples of A1. It is observed that a thermal peak is more distinguished for the dark/brown coloured samples (A1-01-DRK and A2-02-DRK), this suggests a slight difference in composition compared with the clear/colourless samples. Therefore, the dark samples needed more heating time/energy to complete the melting process and reaching a liquid state. The melting point of A1 is found to be 126 °C, and it is matching with a reference low-density polyethene LDPE sample (Resin code #4). Polyethene is used in products such as containers, tapes, and stationary. A low-density polyethene of density equals 0.934 g/cm³ has a melting point of 126 °C, melting heat of 141 J/g, and a degree of crystallisation of 49%. The samples started to experience thermal degradation at a temperature of 220 °C.
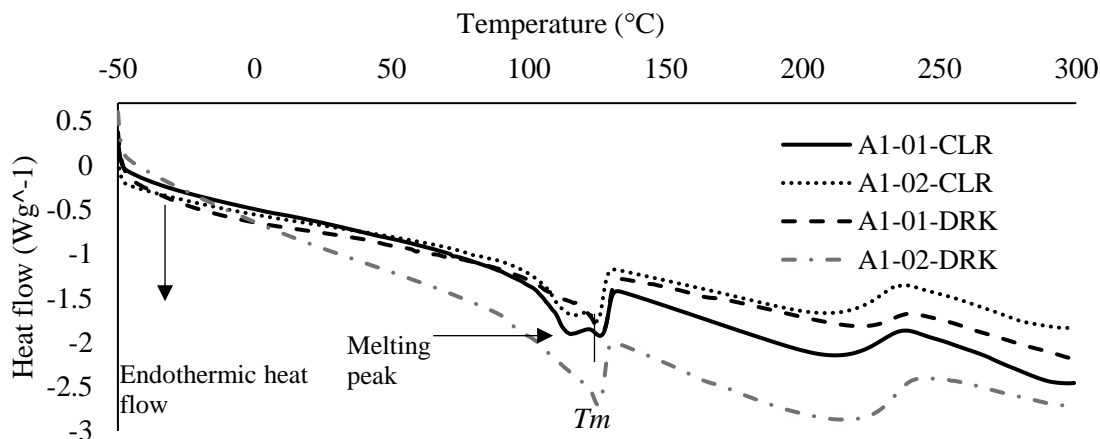

Figure 2: DSC test results for four samples of A1, each has a mass of 5.3 mg placed in open-lid crucibles, tested in nitrogen gas cell at a temperature range of (-50 – 300 °C), with a constant temperature rate of 10 °C/minute. The samples are classified according to their visual appearance, where (CLR) refers to colourless samples, and DRK refers to brown to dark coloured samples.

FT-IR results of A1 are shown in Figure 3 for colourless and dark/brown coloured samples. Generally, the absorption peaks of A1 are identical to the reference absorption peaks of a pure LDPE (2915, 2845, 1467, 1462, 1377, 730 and 717 cm⁻¹). It is observed, however, that there are few extra small peaks for the dark/brown coloured sample, such as at 1741 cm⁻¹, 1578 cm⁻¹ and 1233 cm⁻¹, this can be attributed to the existence of impurities or fillers in low quantities.
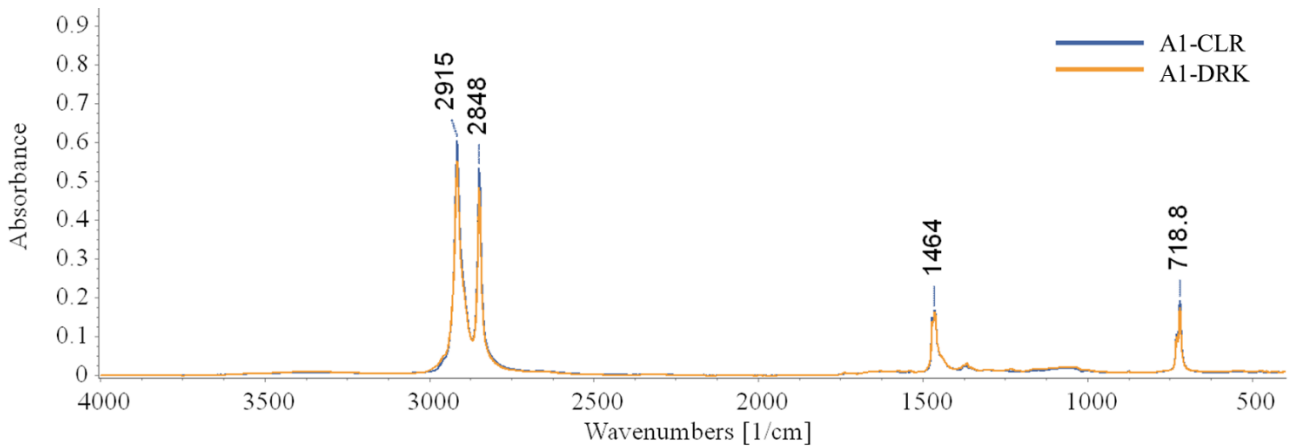
Figure 3: FT-IR test results for plastic-type A1 at a wave range of (400-4000) cm^{-1} showing the absorbance peaks of two samples. A1-CLR indicates the FT-IR result of colourless sample, whereas, A1-DRK indicates the FT-IR result of a dark/brown sample

4.2 DSC and FT-IR results for plastic-type A2

Figure 4 shows the DSC test results for two samples of A2. It is observed that distinctive peaks for A2 samples are directed towards the endothermic heat flow direction, which indicates these peaks (at a temperature range of 165-170 °C) are the melting peaks. When the plastic is heated from its crystal state, the temperature increases at a constant rate, and when it reaches a melting point (*Tm*), the temperature then holds steady until the plastic is completely melted. Once the plastic is melted, the temperature continues to increase again [30], this behaviour is matching with DSC results of A2. There are no other noticeable trends in the DSC results for A2 samples. It indicates that the samples did not experience thermal degradation at the tested heating temperature range (-50 – 300 °C).
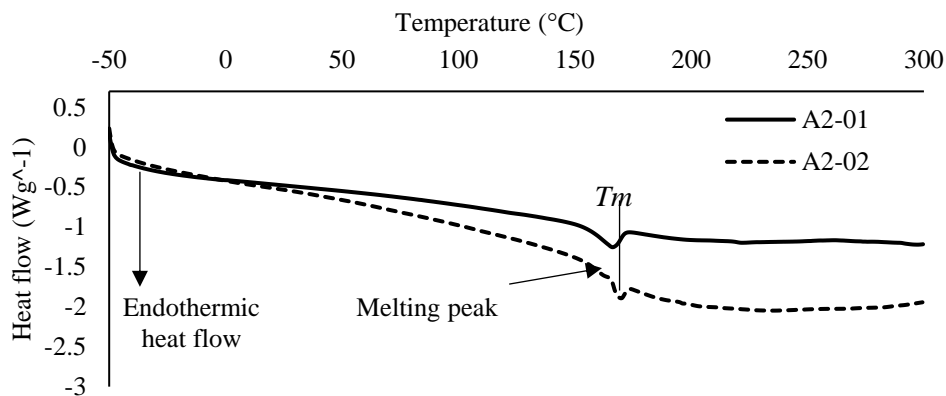
Figure 4: DSC test results for two samples of A2 with samples masses of 5.2-5.3 mg placed in open-lid crucibles, tested in nitrogen gas cell at temperatures range of (-50 – 300 °C), with a constant temperature rate of 10 °C/minute.

FT-IR test result for sample A2 is shown in Figure 5. By comparing the results with reference spectrums of known plastic types, it is found that A2 sample has a similar wave pattern documented for polypropylene (PP) (plastic of Resin code no. 5). PP is a crystalline polymer available at low cost and has excellent formability, resistance to water and high strength. The absorption bands used for the identification of polypropylene are: 2950 cm^{-1} (C-H stretch), 2915 cm^{-1} (C-H stretch), 2838 cm^{-1} (C-H stretch), 1455 cm^{-1} (CH2 bend), 1377 cm^{-1} (CH3 bend), 1166 cm^{-1} (CH bend, Ch3 rock, C-C stretch), 997cm^{-1} (CH3 rock, CH3 bend, CH bend), 972 cm^{-1} (CH3 rock, C-C stretch, CH2 rock, C-CH3 stretch) and 808 cm^{-1} (CH2 rock, C-C stretch, C-CH stretch). It can be observed that few other small peaks could have resulted from the additives and fillers during manufacturing. The melting point obtained by the DSC for A2 (shown in Figure 4) is similar to that of PP polymer (*Tm* range is 160 to 170 °C).
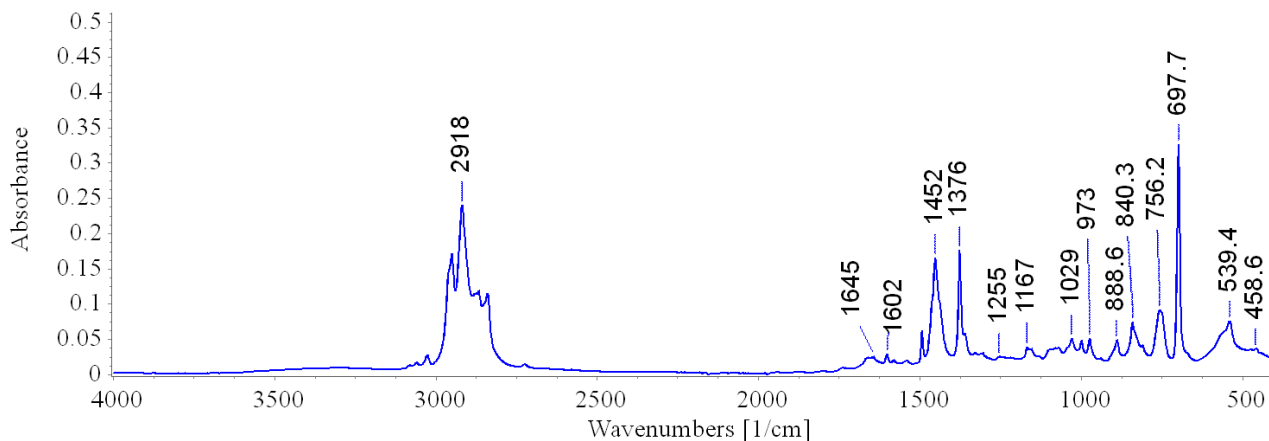
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Figure 5: FT-IR test results for plastic-type A2 at a wave range of (400-4000) cm^{-1} showing the absorbance peaks of the sample.

4.3 DSC and FT-IR results for plastic-type A3

DSC results for two samples of A3 are shown in Figure 6. It can be seen that there are no clear heat flow peaks for A3 at a heating rate of 10 °C/min for a temperature range of (-50 – 300 °C). Therefore, it is difficult to predict this material's composition by using DSC results alone. Since A3 samples did not show a clear melting peak at this temperature range, this may suggest that the plastic could be amorphous polymer. Amorphous polymers are not arranged in crystalline form, but rather chains.
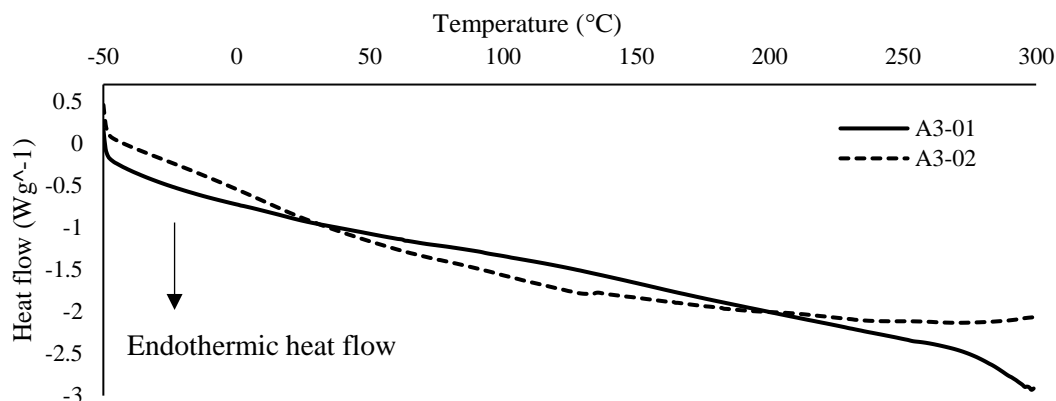
Figure 6: DSC test results for two samples of A3, having masses of 5.4 and 5.3 mg placed in open-lid crucibles, tested in nitrogen gas cell at temperatures range of (-50 – 300 °C), with a constant temperature rate of 10 °C/minute.

FT-IR test result for A3 samples (shown in Figure 7) show absorption bands that correspond to the reference spectrum of poly(vinyl butyral) (PVB). PVB is a strong, transparent and flexible polymer. The absorption bands used for the identification of poly(vinyl butyral) are: 2957.6, 1323.4, 1054.6, 1434.5, 1239.5, 1001.1, 1379.4, 1136.5, 911.5 cm^{-1}. The melting point of the PVB is 90-120 °C. However, the DSC results did not show a clear peak in this range. Therefore, more tests are required to identify the plastic-type used in this recycled material.
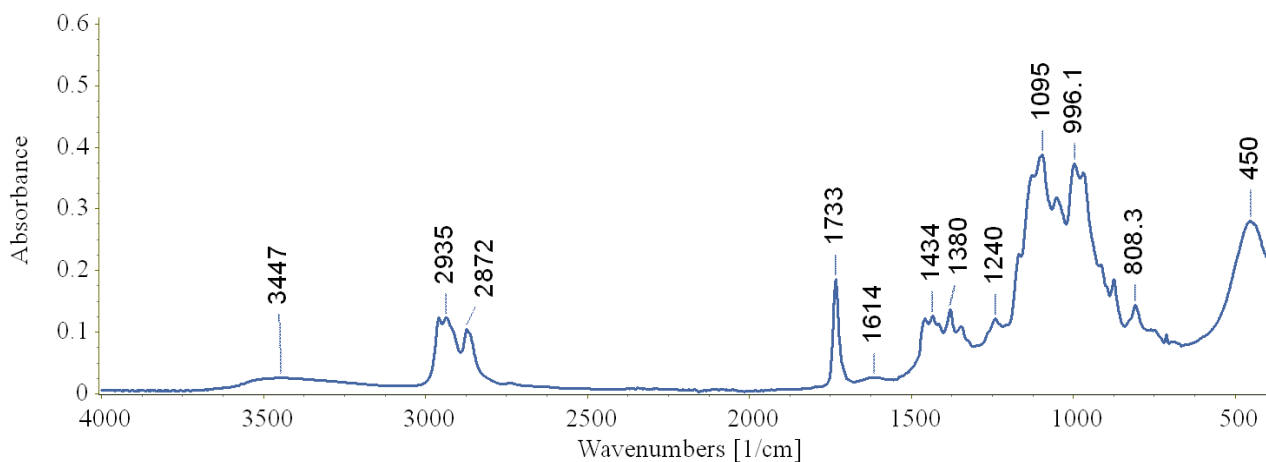
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Figure 7: FT-IR test results for plastic-type A3 at a wave range of (400-4000) cm^{-1} showing the absorbance peaks of the sample.

4.4 DSC and FT-IR results for plastic-type A4

The recycled polymer A4 appears to consist of a variety of materials (see Figure 1). The composition and first use of these materials is unknown. Thus, to identify the thermal properties of A4 polymers by the DSC, mixed components, as well as separate components, are tested. The DSC results are shown in Figures 8a and 8b. Since the material appears to consist of a variety of components, to unify the thermal history of the samples, the samples first heated to a temperature of 190 °C then cooled down to -50 °C and heated again up to 300 °C, using a heating/cooling rate of 10 °C/min.

It is noticed in Figure 8a that the specimen of mixed components yields several melting points; this is reasonable as they consist of different elements, so each component will melt when it reaches a certain heating temperature. The range at which the melting process occurred for all the components is (110 -140 °C), indicating the main plastic type of A4 is LDPE. The samples have also experienced signs of degradation at a temperature of 240 °C. Figure 8b shows the DSC results for three separate, colour-coded specimens collected from A4. It is seen that some samples did not experience a recognisable change in heat flow. Therefore, it is not possible to identify these components by using DSC in the current testing scheme.
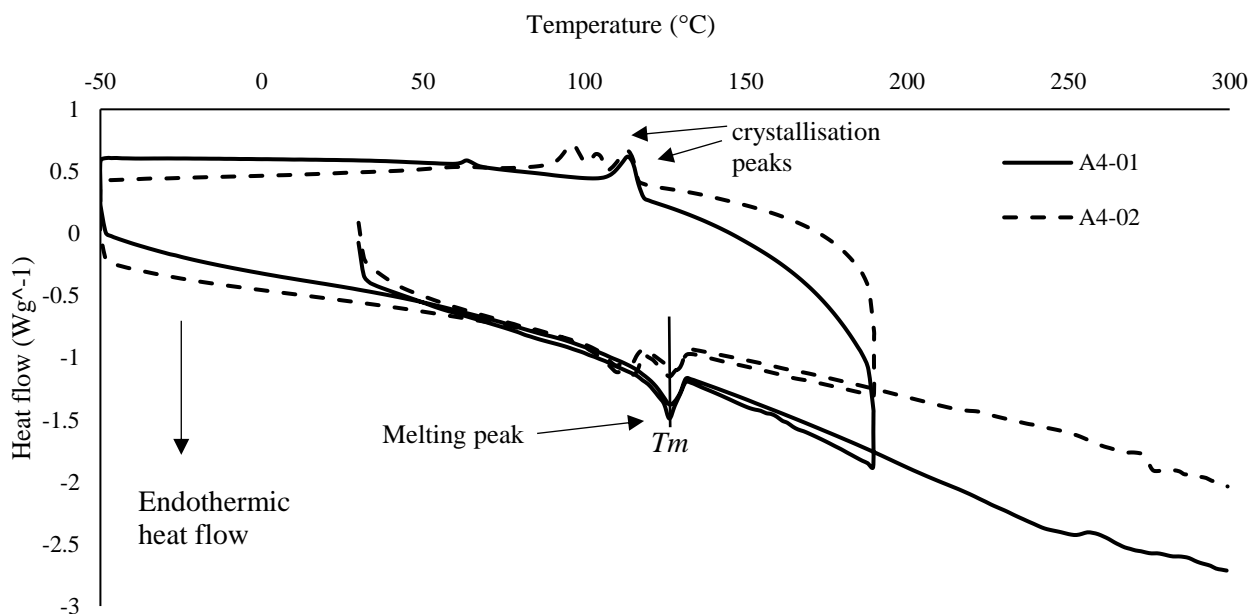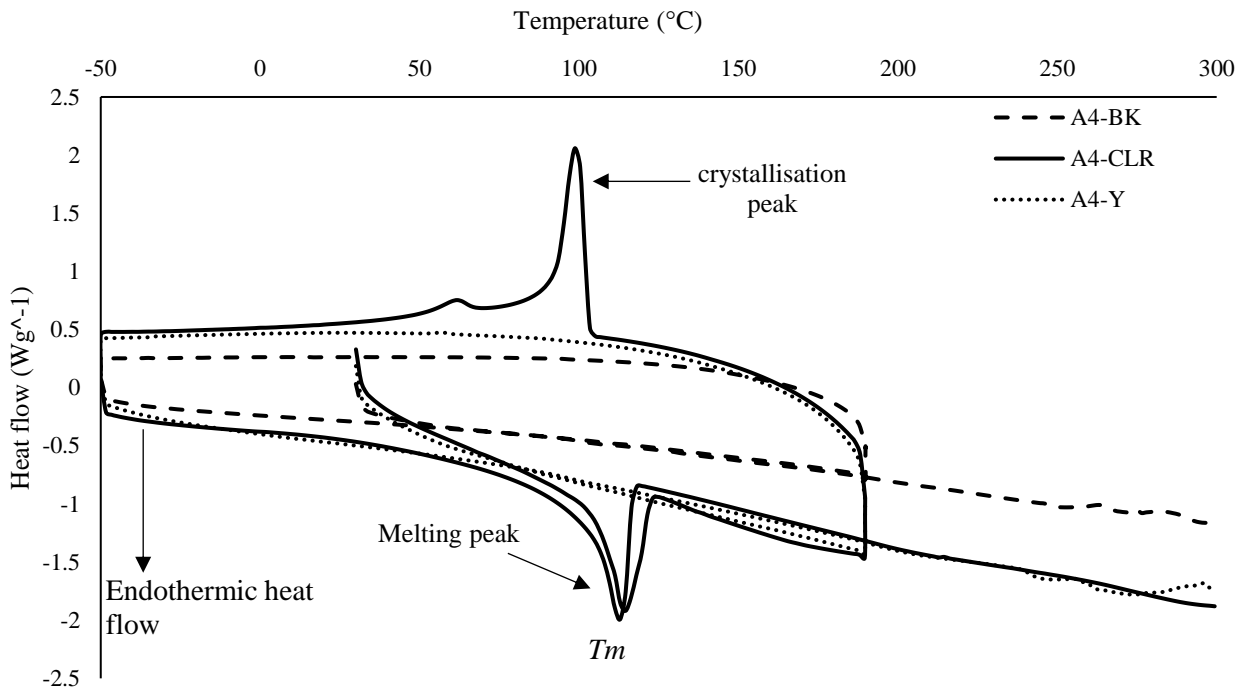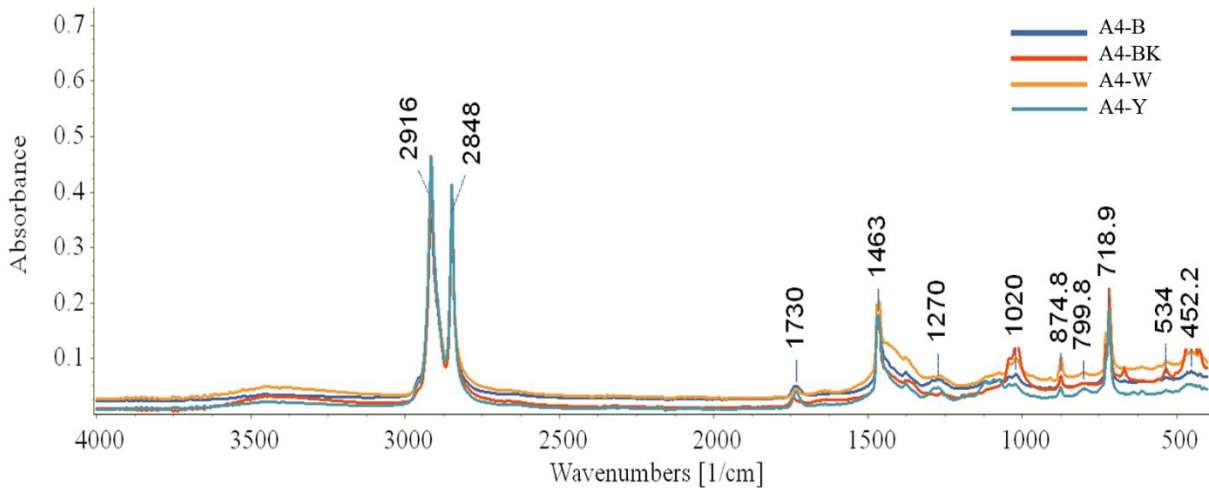

Figure 8a

Figure 8b

Figure 8: DSC test result for the recycled polymer A4, having masses of (5.3-5.8 mg) placed in open-lid crucibles, tested in nitrogen gas cell at temperatures range of (-50 – 300 °C), with a constant temperature rate of 10 °C/minute. The DSC test is conducted for: (a) mixture of A4 components, (b) individual samples selected randomly from A4 plant.

Results of FT-IR scanning of A4 plant are shown in Figures 9-12. The FT-IR results are divided based on similarities in their absorption bands. Accordingly, the results are divided into four groups:

1- Samples A4-B, A4-BK, A4-W, and A4-Y (details for each sample are listed in Table 3) are recognised as LDPE plastics (Resin code #4) (shown in Figure 9a).

Figure 9: FT-IR test results for plastic-type A4 at a wave range of (400-4000) cm⁻¹ showing the absorbance peaks of the samples A4-B, A4-BK, A4-W, and A4-Y

2- Sample A4-CLR (details are listed in Table 3) is recognised as polyethene terephthalate (PETE) (resin code #1). The absorption bands used for the identification of PETE are 1713, 1241, 1094, 720 cm⁻¹. PETE has a high melting point of 260 °C. However, the visual inspection of A4 batch suggests that PETE is not a major component in the sample and can be considered as an impurity. FT-IR results are shown in Figure 10.
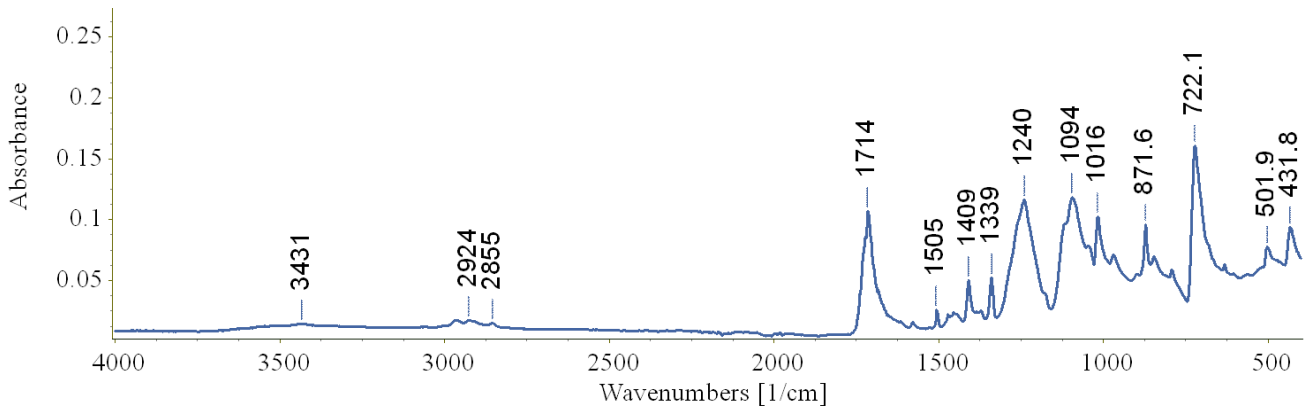
Figure 10: FT-IR test results for plastic-type A4 at a wave range of (400-4000) cm⁻¹ showing the absorbance peaks of the sample A4-CLR

3- Sample A4-G (details are listed in Table 3) is recognised as a combination of polypropylene (PP) (resin code #5) and polyethene (PE). The wavenumbers 2845, 717, 2915 and 1377 cm⁻¹ are used to identify the PE polymer, whereas, the wavenumbers 2950, 2915, 1455, 1377 cm⁻¹ are used to identify the PP polymer (shown in Figure 11).
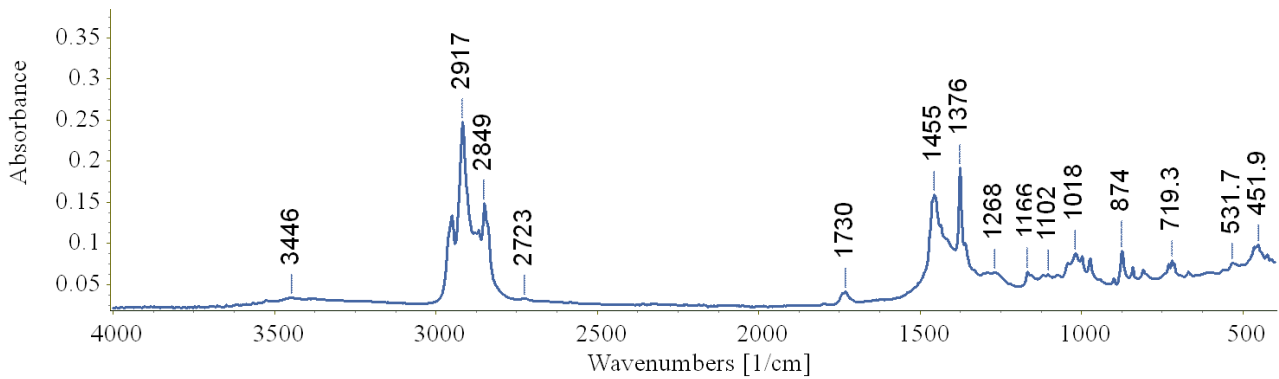

Figure 11: FT-IR test results for plastic-type A4 at a wave range of (400-4000) cm⁻¹ showing the absorbance peaks of the sample A4-G

4- FT-IR test results for samples A4-GY and A4-R (details for each sample are listed in Table 3), shown in Figure 12, show a strong peak at 1724 cm⁻¹ which is related to the ketone group C=O, particularly, the unconjugated ketone. Besides, the cluster peaks at 2920 are all related to C-H stretching, the peaks at 1424 and 1256 are CH3 and CH2, respectively. This functional structure suggests the material belongs to the polyketone group PK. Polyketones are thermoplastic polymers with high resistance to solvents and excellent mechanical properties. The melting point of polyketone is in the temperature range of 220-255 °C.
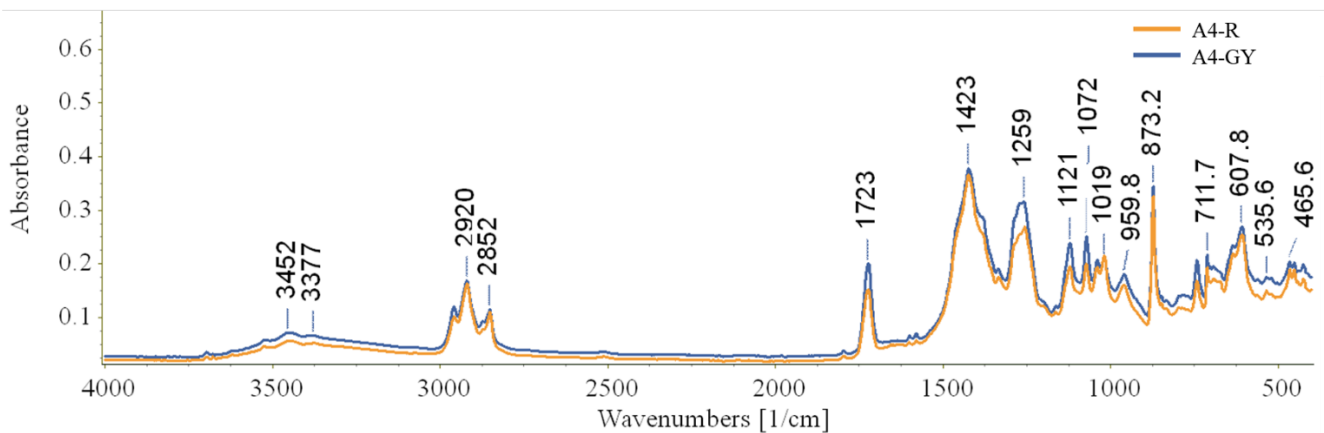

Figure 12: FT-IR test results for plastic-type A4 at a wave range of (400-4000) cm⁻¹ showing the absorbance peaks of the samples A4-R and A4-GY

4.5 DSC and FT-IR result for plastic-types A5 and A6.

Similar to sample A4, the recycled polymers A5 and A6 appear to consist of a variety of materials (see Figure 1). Therefore, to study the thermal properties of these polymers by DSC, specimens of mixed and separate components are tested. DSC results are shown in Figure 13. To unify the thermal history of various components in a sample, the sample is first heated to a temperature of 190 °C then cooled down to -50 °C and heated again up to 300 °C, using a heating/cooling rate of 10 °C/min.

DSC results (Figure 13) show to have a melting point *Tm* at 130 °C, crystallisation peak at 120 °C and start of degradation at 250 °C. The thermal behaviour is found to be identical for all tested A5 and A6 samples. The visual appearance suggests these specimens are manufactured using the same polymer type with different colours. The components of mixed specimens tested in DSC formed one molten mass after heating at 300 °C.
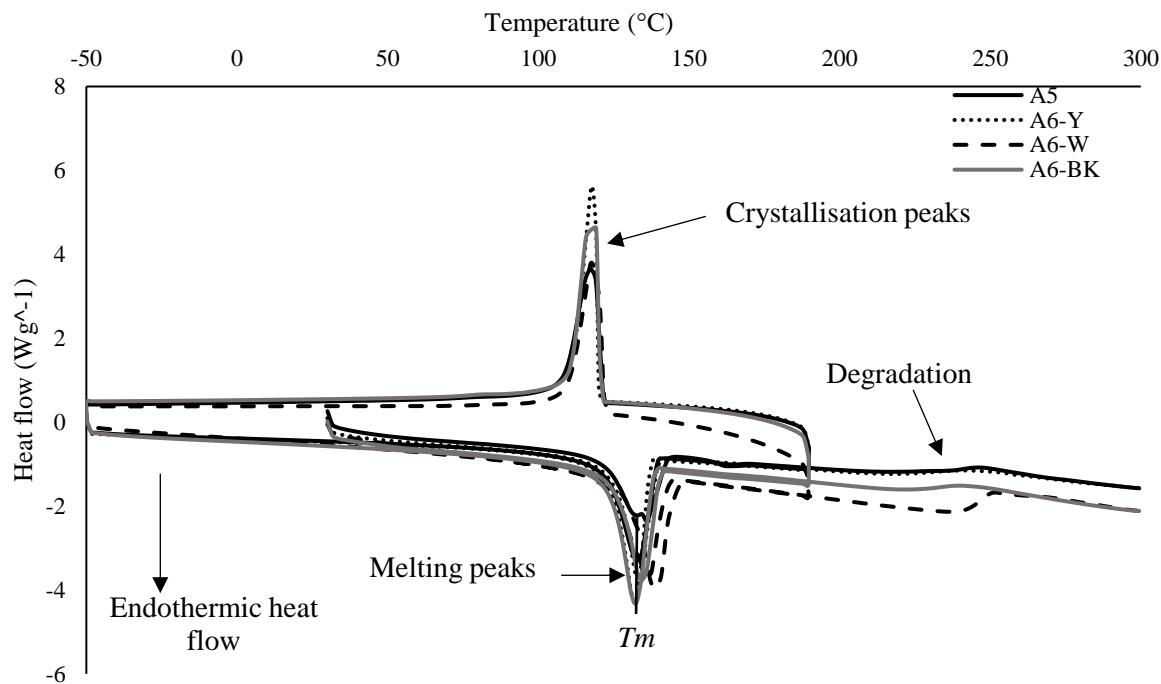

Figure 13: DSC test result for the recycled polymers A5 and A6, having masses of 5.1 to 5.7 placed in open-lid crucibles, tested in nitrogen gas cell at temperatures range of (-50 – 300 °C), with a constant temperature rate of 10 °C/minute.

FT-IR results for A5 and A6 samples (listed in Table 3) are found to be identical (shown in Figure 14); this indicates these materials were manufactured from one type of plastic. By comparing the absorption peaks with a library of reference polymer absorption peaks, it is decided that A5 and A6 are both made of LDPE polymer. The DSC results displayed earlier confirm the same conclusion, that is A5 and A6 plastics consist of LDPE polymer.
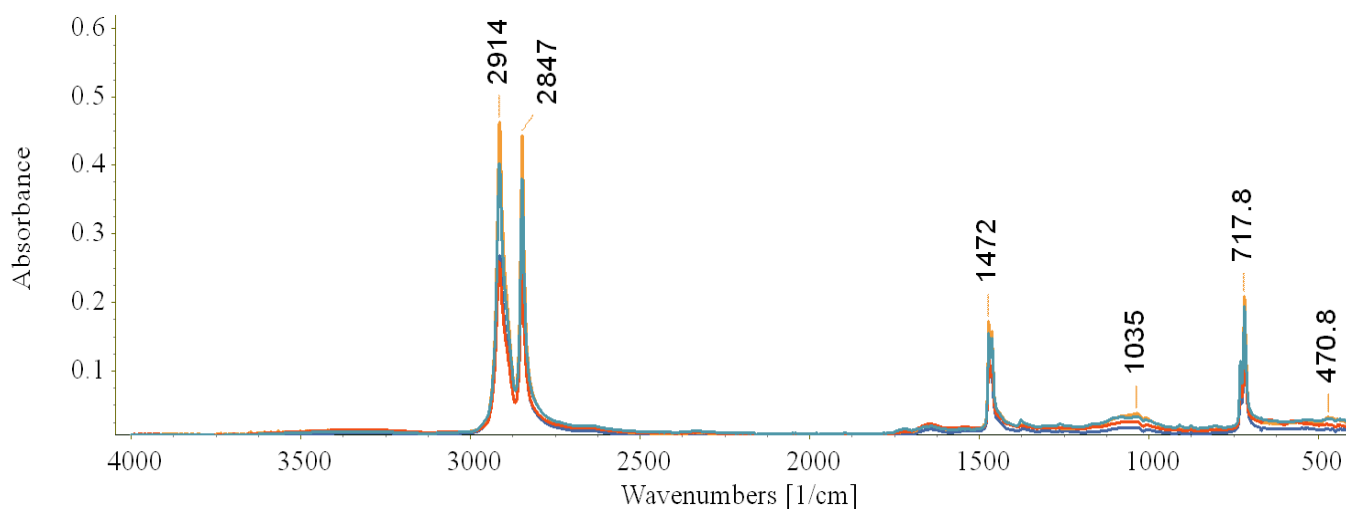
Figure 14: FT-IR test results for multiple samples from plastic-types A5 and A6 at a wave range of (400-4000) cm^{-1} showing the absorbance peaks.

5 RHEOLOGICAL AND MECHANICAL CHARACTERISATION OF WASTE PLASTICS-MODIFIED BITUMEN

FT-IR and DSC tests are conducted to identify the plastics types and thermal properties of tested recycled waste plastics. It is found that the materials named A1 and A2 consist of low-density polyethene (LDPE) and polypropylene (PP), respectively. Other recycled plastics (A3 and A4) consist of a variety of materials and impurities. For this reason and because A1 and A2 are completely characterised and found to possess a melting point lower than 170 °C which can be reached in the pavement mixing plants without significant alteration to standard practices, A1 and A2 are selected in the current study to modify the bitumen binder. Because the recycled waste plastics (A5 and A6) also consist of low-density polyethene, they are not considered in the study to avoid duplication of the materials.

The control bitumen X70 is mixed with 6 % of A1 and A2, and the mixture is characterised by using DSR testing schemes to measure the rheological properties and the engineering performance of the waste plastic-modified bitumen. Results are discussed in the following sub-sections.

5.1 Viscoelastic properties of the waste plastic-modified bitumen

The frequency sweep test determines the degree of viscosity and elasticity in the modified bitumen binders at undamaged loading conditions. A temperature range of 10-70 is selected, and a loading frequency of 0.1-25 Hz is set. Figures 15 (a, b and c) show the phase angle versus loading frequency for the control binder X70, and A1- and A2-waste plastic-modified binders, respectively. It can be observed that the addition of 6% A1 and A2 causing the phase angle to decrease compared to the control sample. Therefore, the waste-plastic is acting more towards the elastic material. This trend of the waste plastic-modified bitumen to have more elastic behaviour is somehow expected when tested at a temperature lower than the melting point for these plastic compounds.

Figure 16 shows the TTSP generated master curves for the complex shear moduli of the control bitumen X70, A1- and A2-waste plastic-modified samples. The shear modulus of A1-modified bitumen experienced less sensitivity to frequency and a higher shear modulus at low frequencies compared with the control sample X70. The shear modulus experienced a slight increase for A1-modified bitumen, while, the phase angle experienced a small drop. Therefore, it is expected that A1-modified bitumen will have higher rutting resistance (G*/sin δ) and stiffer behaviour compared to the control binder. A2-modified bitumen sustained a similar shear modulus-frequency relationship to that for X70 control sample but with overall slightly lower stiffness represented by a general decline in the shear moduli. The reason for the slight drop in the shear modulus of A2-modified bitumen while maintaining a lower phase angle compared to the control binder is unclear. Subsequently, the effect of this change on the fatigue, rutting and healing of bitumen is unclear and will need further testing. Nonetheless, it can indicate a lower resistance to fatigue (by considering the fatigue parameter G* sin δ). Previous studies [14] proposed the inclusion of PP plastic to bitumen will work as an elastomeric additive, expecting to achieve increased recovery capacity, but less resistance to deformation. Therefore, the following sections will further investigate these performance properties.

(a) X70 control bitumen　　**(b) A1-modified bitumen**
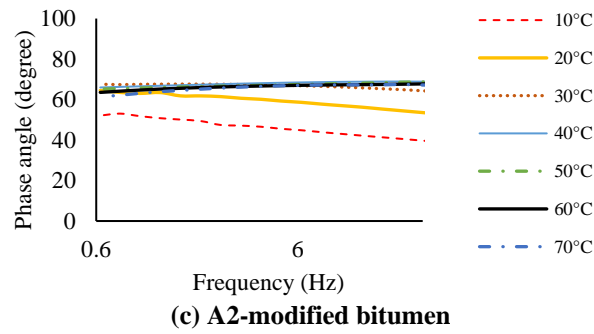
(c) A2-modified bitumen
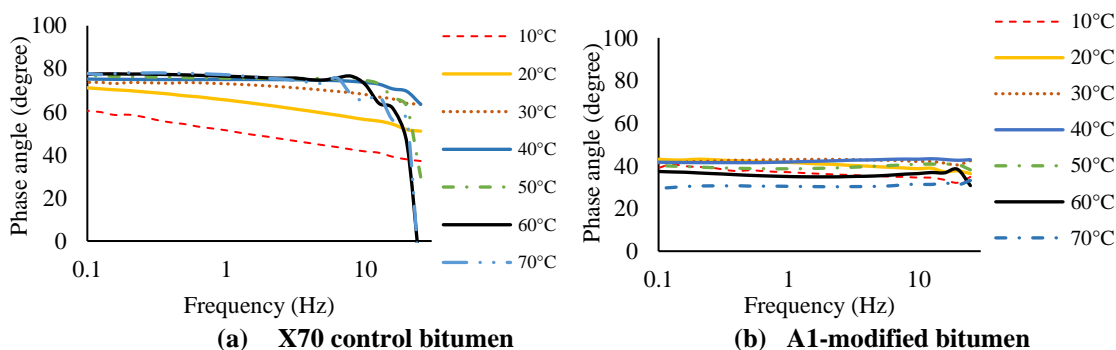
Figure 15: Frequency sweep test results for (a) X70 control bitumen; (b) A1-modified bitumen and; (c) A2-modified bitumen. The test is conducted at temperatures of 10 to 70 °C with a loading frequency of 0.1-25 Hz.
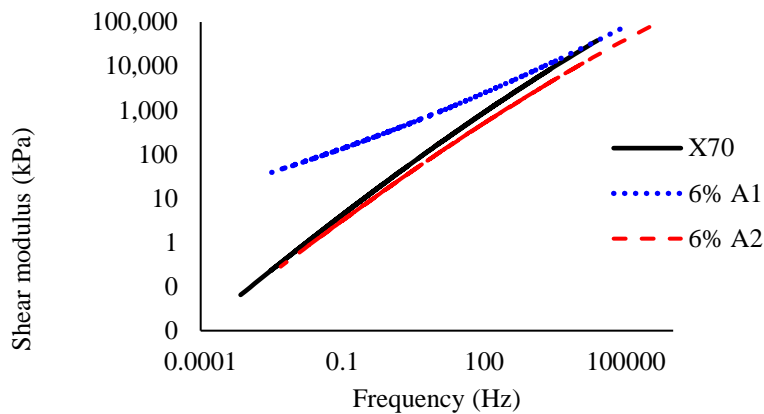

Figure 16: Shear modulus master curves calculated using the time-temperature superposition principle for the frequency sweep test for X70 control bitumen sample, A1- and A2-modified bitumen samples.

5.2 Fatigue-resistance performance

In order to evaluate the fatigue performance effectively, DSR records the viscoelastic parameters (i.e. shear modulus and phase angle) with the progression of time in the TS testing scheme. By selecting the appropriate conditions for the DSR parameters, the fatigue performance can be examined. In this study, a strain level of 5%, a frequency of 10 Hz and an intermediate temperature of 20 °C are set. Figure 17 shows the TS results for A2-modified bitumen. The shear modulus shows a sharp initial decrease until it stabilises with a slow rate of decline following the initial stage. This behaviour can be attributed to the fatigue cracking stages that the sample experience when subjected to fatigue load, where the initial stage (e.i., sharp drop in shear modulus) is caused by the 'edge flow' occurring in the sample [31, 32].
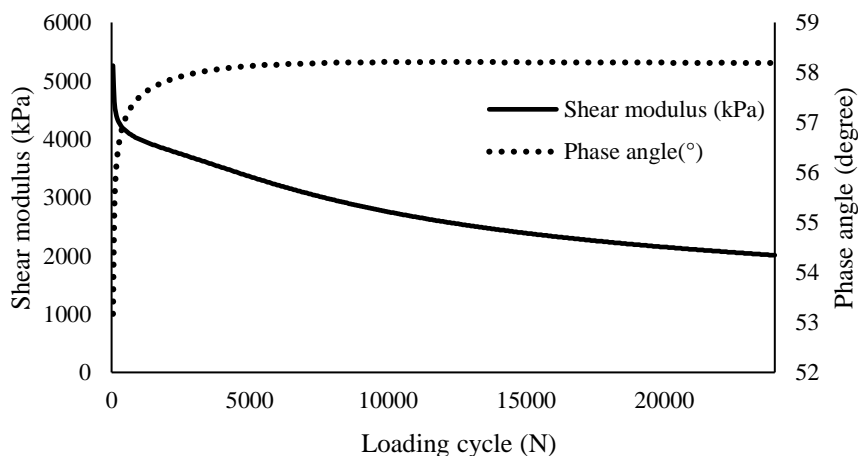
Figure 17: Time sweep (TS) test results for A2-modified bitumen (bitumen with 6% A2). The test is conducted at 20 °C temperature, 10 Hz loading frequency, 5% strain level, and up to 24,000 loading cycles.

Figure 18 illustrates the shear modulus results of the bitumen samples X70, A1- and A2-modified bitumen obtained from TS tests. A2-modified bitumen and control samples both have similar initial shear modulus, so this enables cross-comparison. However, the reduction in shear modulus as the loading cycle increase is more prominent with the control bitumen X70. More specifically, X70 bitumen showed a faster decrease in shear modulus, while A2 shows a gradual regression. This behaviour suggests the addition of A2 to bitumen is useful to improve the fatigue cracking resistance because it results in a lower energy dissipation through the crack formation. Hence, a lower percentage of damage accumulates. Whereas, the inclusion of A1 exerts the steepest decline in comparison to the control and A2-modified bitumen. It becomes challenging to determine the actual difference between shear modulus of A1-modified sample and control bitumen because a considerable variation exists in the initial shear modulus.
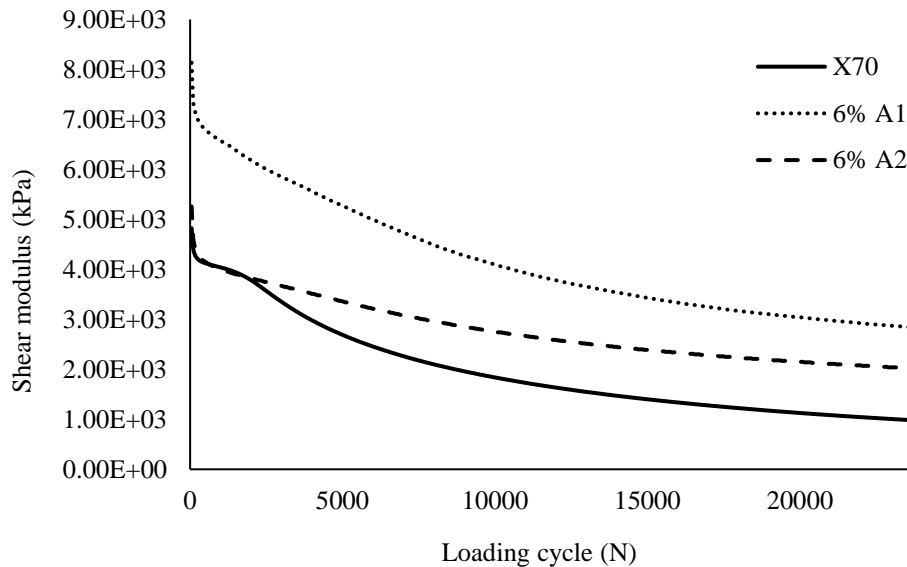

Figure 18: Time sweep test results showing the shear modulus vs loading cycle for bitumen X70, and A1- and A2-modified bitumen samples.

Figure 19 represents the phase angle obtained from the TS test for X70 bitumen, and A1- and A2-modified bitumen samples. The A2-modified sample has a lower phase angle across the loading cycles and therefore, a greater ability to resist fatigue damage in comparison to the control sample X70. With A1-modified sample, it becomes difficult to compare as the phase angle during the initial loading cycle is significantly different. The initial phase angle value for A1-modified sample is 43.24 °, but for X70 bitumen it is 58.19 °. Nevertheless, a low initial phase angle value implies a better fatigue performance. Generally, there is an improvement in the fatigue resistance with the addition of plastic extenders.
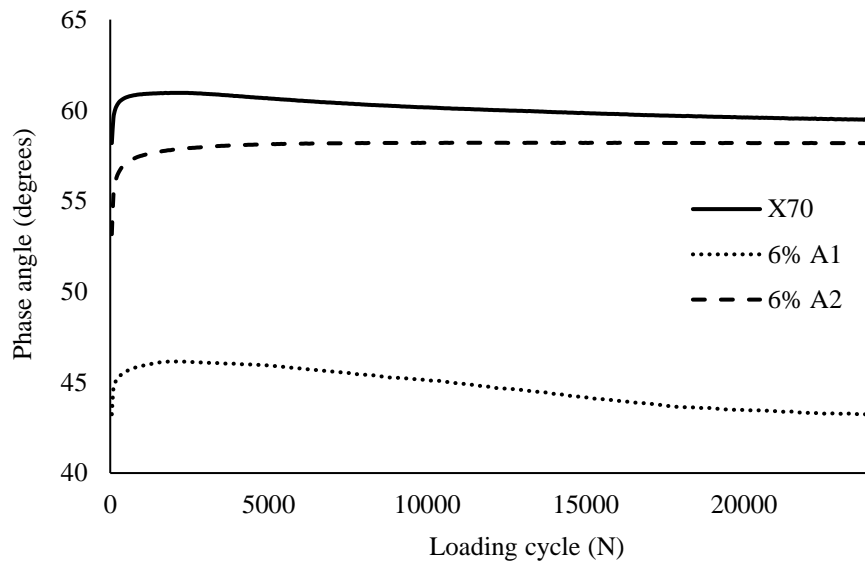
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Figure 19: Time sweep test results showing the phase angle versus loading cycle for bitumen X70, A1-, and A2-modified bitumen

Time sweep test results are utilised to obtain the DSR-Crack Growth (DSR-C) graph. The crack length of asphalt binders subjected to rotational shear are predicted in DSR-C method (shown in Eq. 1). Undamaged and damaged conditions measure the crack growth and the extent of fatigue damage undertaken by the TS test for all three binder samples. Figure 20 shows the crack length development with the loading cycles for all tested samples (i.e., X70, 6% A1-modified bitumen, and 6% A2-modified bitumen). There is a significant difference in the crack length for X70 bitumen and plastic modified samples with the latter having smaller crack lengths. This indicates binders with 6% A1 and 6% A2 sustained low susceptibility to cracking, and thus, A1 and A2 can prolong the fatigue life of the pavement. Furthermore, cracking length for A1- and A2-modified samples are very similar, suggesting they acquire similar fatigue resistance properties. It is also observed that at a lower number of loading cycles ($n < 3000$ loading cycles) the initial crack length is higher for A1- and A2-modified samples. This initial cracking behaviour, if considered alone (i.e., by examining the results of undamaged TS test or frequency sweep test) can be misleading.
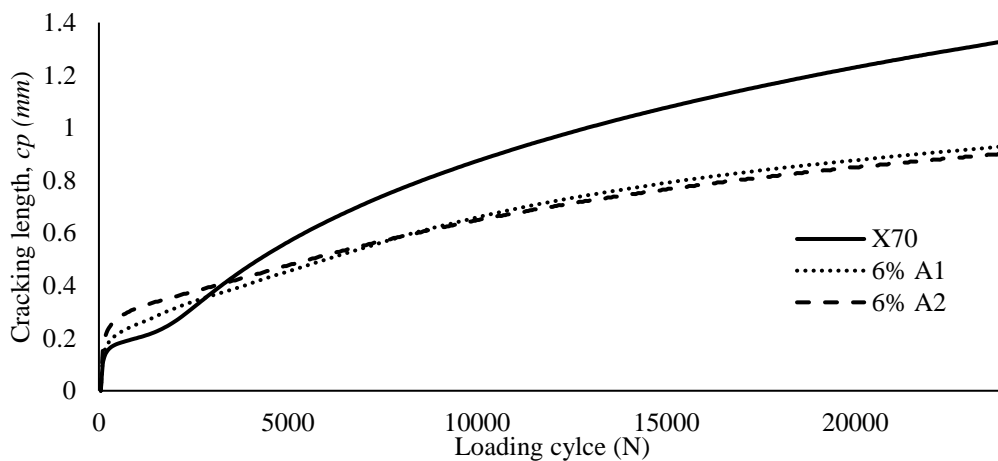

Figure 20: The crack length propagation under time sweep (TS) test conditions for bitumen X70, A1- and A2-modified bitumen. The crack lengths are calculated using DSR-C method.

5.3 Self-healing performance

Figure 21 demonstrates the healing results of all tested samples, in which the DSR specimens are subjected to two constitutive time sweep tests separated by a rest (recovery) period. This DSR-testing scheme characterises self-healing of bitumen as the recovery of the stiffness when the binder is inclined to damage created by cyclic fatigue loading. It can be observed from Figure 21 that the samples experienced a partial recovery in the second run of TS tests, and the second run of results have a less defined steepness in the curve in comparison to the first run.
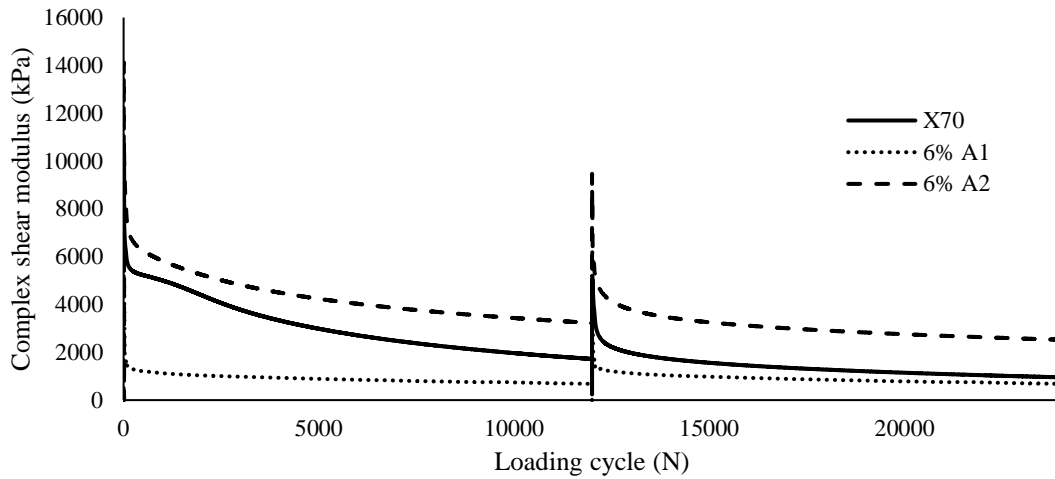
Figure 21: Healing test results showing the shear modulus versus loading cycles for control bitumen sample X70, and A1- and A2-modified bitumen samples

To quantify the healing potentials, the difference in shear moduli before and after a rest period can be further observed for tested asphalt binders. The healing ability of the control bitumen X70, and A1- and A2-modified bitumen specimens are evaluated using the Healing Index (HI), illustrated in Eq. 2. A better healing potential will have a higher HI value. The healing indices for all three samples are found to be 65.80, 65.86 and 88.61 for A2-modified bitumen, X70 control bitumen, and A1-modified bitumen, respectively. This suggests that some recycled waste plastics possess greater healing potential (i.e., LPDE) while some do not (i.e., PP), in comparison to the base binder.

To conclude, LDPE-recycled plastic has a better built-in capability to recover microcracks; therefore, it can improve the durability of the bitumen binders. PP-modified bitumen had almost an identical healing potential to that of the original binder X70. Therefore, using recycled waste plastic A2 to modify X70 bitumen does not enhance its fatigue-healing efficiency.

5.4 Rutting resistance

MSCR test is conducted on the control bitumen sample X70, and A1- and A2-modified binders at 60 °C. The non-recoverable compliance (J_{nr}) and percentage recovery are plotted against all tested samples for comparison in Figures 22 and 23.
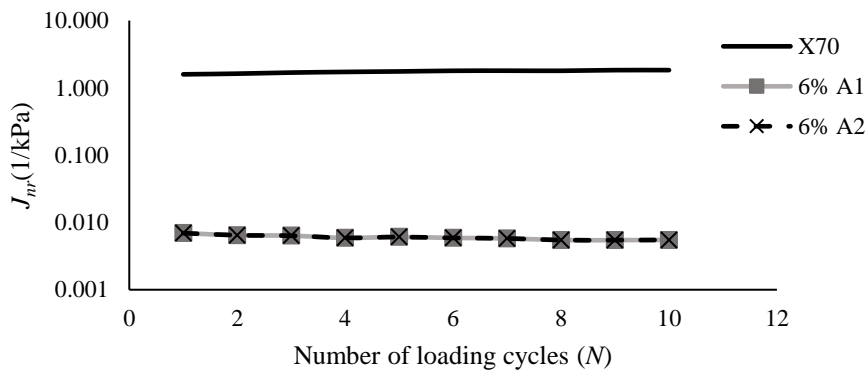

Figure 22: J_{nr} results for bitumen sample X70, A1- and A2-modified bitumen samples

It can be observed from Figure 22 that A1- and A2-modified bitumen acquire very similar J_{nr} values, so there is a similar expectation for their rutting performance. The J_{nr} values for both recycled waste plastics suggests that the plastic-modified binders can lead to a significant improvement in high-temperature rutting performance in comparison to X70 as the J_{nr} values are comparably lower.

Figure 23 outlines the percentage recovery of the control bitumen sample X70, and A1- and A2-modified bitumen samples. The percent recovery indicates the delayed elastic response of an asphalt binder; therefore, it is desirable to obtain a higher percent recovery to reduce the susceptibility for rutting failure. From the examination of results, it s observed that the recovery of X70 bitumen is relatively poor. Whereas, the plastic-modified binders have a substantially improved rutting resistance as there is an enhancement of the % recovery. The recovery has improved

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by 8% and 24% for both A1- and A2-modified binders, respectively across the loading cycles 1 to 10 N, thus supporting the above trend.
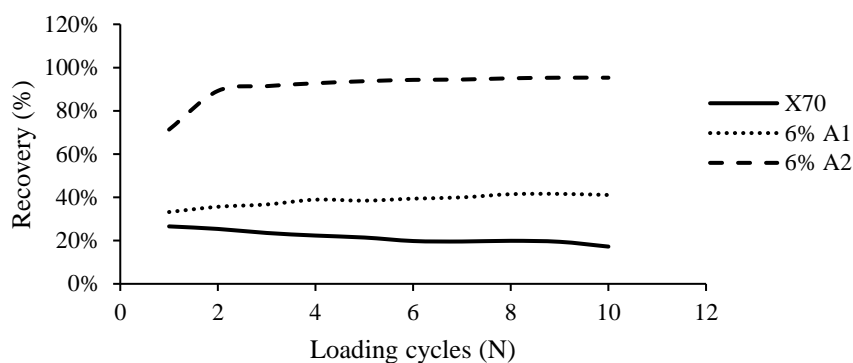

Figure 23: Percentage recovery for bitumen samples X70, and A1- and A2-modified bitumen samples

6   Conclusions

Incorporating plastics in the manufacturing of flexible pavements helps consume large quantities of plastic wastes and reduce the demands for producing petroleum-based bitumen. Therefore, the current study investigates the chemical and physical characteristics of the plastics waste produced by local waste recycling manufacturers, and measure their impact on the bitumen binder when mixed. Six waste-plastic types which are produced in great quantities by a local waste plastic-recycling supplier are tested. Then, two classes are nominated for bitumen modification. A series of rheological and mechanical tests are conducted on the waste-plastic modified bitumen. The conclusions are summarised as follows:

1- The materials named A1 and A2 consist of low-density polyethene (LDPE) and polypropylene (PP), respectively. Other recycled plastics A3 and A4 found to consist of a variety of materials and impurities. A1 and A2 are selected in the current study to modify the bitumen binder, as they have a melting point lower than 170 °C which can be reached in the pavement mixing plants without significant alteration to the standard practices. A5 and A6 are also found to consist of low-density polyethene. For this reason, they are not mixed with bitumen in this study to avoid duplication of materials. However, they can also be utilised as binder extenders.

2- The plastic-modified samples examined in this study have significantly altered the viscoelastic behaviours of the bitumen binders. A1-modified bitumen (mainly consists of LDPE) exhibited more elastic and less viscous performance compared to the control bitumen, showed by increased shear modulus and reduced phase angle. A2 (mainly consists of PP) reduced the shear modulus significantly for the control bitumen.

3- The plastic-modified binders (A1 or A2-modified bitumen) both have a substantially improved rutting resistance as there is an enhancement of the % recovery and a reduction in the non-recoverable strain compliance J_{nr}. In comparison, A1 led to an increased healing potential to the bitumen while the A2 did not show obvious improvement to the healing performance of the bitumen.

4- The addition of plastic modified bitumen has improved fatigue performance. Cracking lengths of waste plastic-modified bitumen are much lower compared to that for the control bitumen. Thus, they exhibit low susceptibility to cracking, therefore helping to prolong the pavement's fatigue life. The two waste plastic-modified bitumen samples had similar cracking lengths, suggesting they acquire a similar fatigue resistance.

This study covered the thermal and chemical characterisation of several locally-produced recycled waste plastics for asphalt pavements. Preliminary mechanical performance characterisation of two waste plastic-modified bitumen at the unaged condition is compared. Further tests are conducted to examine the field performance of waste plastics-modified bitumen and mixtures. Generally, using recycled PP and PE materials in the pavement industry is promising.

ACKNOWLEDGEMENTS

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