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Rheological and MECHANICAL Performance of Asphalt Binders and Mixtures Incorporating CaCO₃ and LLDPE

Mohd Rosli Mohd Hasan^{1*} Zhanping You^{2*} Mohd Khairul Idham Mohd Satar³ Muhammad Naquiuddin Mohd Warid³ Nurul Hidayah Mohd Kamaruddin⁴ Sharvin Poovaneshvaran¹

1. School of Civil Engineering, Universiti Sains Malaysia, Engineering Campus, 14300 Nibong Tebal, Pulau Pinang, Malaysia

2. Department of Civil and Environmental Engineering, Michigan Technological University, 1400 Townsend Drive Houghton, MI 49931, USA

3. Department of Geotechnics and Transportation, Faculty of Civil Engineering, Universiti Teknologi Malaysia, 81310 Johor Bahru, Johor, Malaysia

4. Faculty of Civil and Environmental Engineering, Universiti Tun Hussein Onn Malaysia, 86400 Parit Raja, Batu Pahat, Johor, Malaysia.

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ABSTRACT

This study was conducted to assess the performance of modified asphalt binders and engineering properties of mixtures prepared with incorporation 3 vol% and 6 vol% of calcium carbonate (CaCO₃), linear low-density polyethylene (LLDPE), and combinations of CaCO₃ and LLDPE. The rheological properties of control and modified asphalt binders were evaluated using a series of testing such as rotational viscometer (RV), multiple stress creep recovery (MSCR) and bending beam rheometer (BBR) tests. Meanwhile, four-point beam fatigue test, the dynamic modulus (E*) test and tensile strength ratio (TSR) test were conducted to assess the engineering properties of asphalt mixtures. Based on the findings, the RV and MSCR test result shows that all modified asphalt binders have improved performance in comparison to the neat asphalt binders in terms of higher viscosity and improved permanent deformation resistance. A higher amount of CaCO₃ and LLDPE have led modified asphalt binders to better recovery percentage, except the asphalt binders modified using a combination of CaCO₃ and LLDPE. However, the inclusion of LLDPE into asphalt binder has lowered the thermal cracking resistance. The incorporation of CaCO₃ in asphalt mixtures was found beneficial, especially in improving the ability to resist fatigue cracking of asphalt mixture. In contrast, asphalt mixtures show better moisture sensitivity through the addition of LLDPE. The addition of LLDPE has significantly enhanced the indirect tensile strength values and tensile strength ratio of asphalt mixtures.

**Corresponding Author:*

Zhanping You,

Department of Civil and Environmental Engineering, Michigan Technological University, 1400 Townsend Drive Houghton, MI 49931, USA;

Email: zyou@mtu.edu ;

Mohd Rosli Mohd Hasan,

School of Civil Engineering, Universiti Sains Malaysia, Engineering Campus, 14300 Nibong Tebal, Pulau Pinang, Malaysia;

Email: cerosli@usm.my

1. Introduction

The most critical issue and the challenge of asphalt pavement in the cold regions is thermal or low-temperature cracking. Though, the performance of asphalt pavement at high temperature is similarly crucial to take into consideration to ensure long service life. As a visco-elastic material, the performance of bituminous material heavily depends on both temperature and time^[1,2]. The modulus of asphaltic concrete pavement can be varied; high during winter conditions and low in hot summer days^[3]. The visco-elastic characteristics of asphalt binder enable the asphalt binder to flow at elevated temperature, and it also can withstand a long loading time. Anyhow, due to its elastic characteristics that present in the asphalt binder, the asphalt binder tends to become brittle at a reduced temperature as well as only able to sustain short loading time. The brittle characteristics of asphalt binders during low temperature could result in thermal cracking failure which would be the principal reason to the premature pavement problems. Contrarily, rutting commonly develops during the hot seasons, when the asphalt is softer. Asphalt binder consists of a complex chemical mixture, which mainly differs in size and components of hydrocarbons, as well as different elements in the functional group, such as nitrogen, hydrogen, and sulphur atoms^[4]. The asphalt material also consists of organic molecules, where the molecules can react with oxygen from the surrounding area. The extent of ageing and oxidation reactions are greatly influenced by temperatures, types of modifier and concentration, which could make the binder stiffer and brittle throughout the time. Aging and oxidation reactions significantly affect the short and long-term performance of pavement materials^[5-8].

Over the years, polyethylene (PE) resin had been utilized in pavement material to modify the performance of asphalt binders and mixtures^[9-14]. The usage of polyethylene and polypropylene as binder modifiers could enhance the resistance to permanent deformation under traffic loading since both of this materials are characterized as plastomers, which can increase the stiffness of the modified binders^[9,10]. Polacco et al.^[9] reported that the PE related modifiers were strongly biphasic, where the material tended to separate into asphalt-rich and polymer-rich phases. However, LLDPE demonstrated greater compatibility with asphalt, which strongly improved the mechanical properties of modified asphalt binder in comparison to the base asphalt binder. The presence of LLDPE modifier in the asphalt binder has a substantial reaction with the asphalt through a continuum matrix of the polymer chains. The polymer continuum matrix permitted the modified

asphalt to exhibit a polymer-like behaviour even though insufficient polymer content. Besides that, Drozdov and Yuan^[15] specified that the morphology of polyethylene is influenced by a certain degree of stress and deformation due to the movement of chains that occurred at the nodes. Fang et al.^[14] concluded that additions of LLDPE waste had improved the softening point of the modified asphalt binder from 47 °C to 76 °C, which lead to a reduction in ductility and penetration values. These results indicated that LLDPE has significantly improved the high-temperature behaviour and viscoelasticity of bitumen.

Furthermore, the author stated that the LLDPE is denoted by a multi-branching structure comprising of a long chain of linear molecules with alkyl and methyl-branched chains. These two materials are not thermodynamically incompatible; where the materials were not completely dispersed at the molecular level, due to delimitation between the materials. Through this combination, the individual components of asphalt reacted with LLDPE molecules chains causing them to swell and link together. It allowed the modified asphalt binder to absorb more energy and improve performance at high temperature^[16,17].

Limestone or calcium carbonate (CaCO_3) is an inert material that came into regular use as an additive in asphaltic concretes in the 1980s^[18]. The powder form of hydrated lime exhibits higher surface area which contributes towards rapid chemical reaction. The ions present in the interface of aggregates and asphalt binder electrically repelled each other since both the material comprises acids. In order to improve the adhesion bonding of both materials, alkali in the form of hydrated lime was added to neutralize the acidic conditions by introducing the opposite-charge ions that could strengthen the adhesion bonding^[19]. Additionally, the usage of lime in asphalt mixture is not only limited as an anti-stripping agent, but it may also be used to increase the asphalt mixture stiffness, reduce plasticity index if clays exist and reduce the oxidation rate^[18,19]. However, the amount of CaCO_3 and the proper preparation method to be used is critical to ensure the effectiveness of improving the performance of asphalt materials^[20].

The two approaches that are typically used to add polymers to the asphalt binder, which are dry and wet methods. In general, the dry method can be done by adding the polymer directly into the aggregate, before incorporating the binder during the asphalt mixture preparation process. This process requires an adequate mixing duration to ensure the polymer homogeneously disperses in the asphalt mixture. However, in regards to the wet process, the polymer material is initially mixed with the asphalt binder at designated temperature prior to mixing with the aggregate.

The mixing temperature and duration as well will depend on the type of asphalt binder and polymer material ^[21,22].

The scope of this study covered investigations on the bitumen behavior and the engineering properties of asphalt mixtures prepared with the addition of different amounts of CaCO₃ powder, LLDPE plastic, and modifier contained the combination of both LLDPE and CaCO₃. The rheological properties of asphalt binders were evaluated based on the standard Superpave specification tests. These include volatile loss test, rotational viscometer (RV) test, dynamic shear rheometer (DSR) test and bending beam rheometer (BBR) test. Then, the asphalt mixture specimens were assessed using the four-point beam fatigue test, the dynamic modulus (E*) test, and modified Lottman test. During the sample preparation, the wet process was adopted, where the modifier was mixed with bitumen at designated temperature prior to use in the asphalt mixture preparation.

2. Materials

2.1 Asphalt Binder and Aggregate

The main materials used in this study were obtained from Payne & Dolan Inc, located in Hancock, Michigan. The PG 58-28 was selected as a control binder. The aggregate gradation adopted for the preparation of asphalt mixtures was based on the Michigan Department of Transportation (MDOT) as shown in Table 1. The course and fine aggregates used in this study were basalt and natural sand, respectively. The nominal maximum aggregate size is 12.5 mm, and the designed traffic level is less than 3 million equivalent single axles loads (ESALs) based on the current Superpave asphalt mixture design practice.

Table 1. Aggregate gradation for the preparation of asphalt mixtures

Sieve Size (mm)	Percent Passing (%)
19.00	100.0
12.50	94.0
9.50	86.3
4.75	68.2
2.360	49.2
1.18	38.4
0.60	27.8
0.30	15.0
0.15	6.7
0.075	4.5
Pan	0.0

2.2 Linear Low-Density Polyethylene

The Dow Chemical Company supplied the linear low-density polyethylene (LLDPE) pellet resin as shown in Figure 1(a) was used in this study. The physical appearance of the pellets is white in colour and odourless ^[23]. The density of the pellets is 0.917 g/cm³. It was used as-received.

2.3 Calcium Carbonate

Ground calcium carbonate (CaCO₃, uncoated) was used as a filler material in the production of asphalt modifiers. The CaCO₃ (Figure 1(b)) used was supplied by Specialty Minerals Inc. ^[24]. The material is characterized by superior brightness, and controlled particle size (mean particle size = 3.5 μm, +325 mesh = 0.004 wt%). The CaCO₃ consists of 97 wt% CaCO₃, 0.5 wt% Magnesium Carbonate (MgCO₃), less than 0.1 wt% Iron as (Fe₂O₃), and less than 0.2 wt% moisture content ^[25]. The specific gravity of this material is 2.7, which is higher compared to other modifiers used in this study. The CaCO₃ was used as-received and has a surface area of 3.1 m²/g.

2.4 Polyethylene-Calcium Carbonate (PECC) Pellets

PECC pellet (Figure 1(c)) was produced by combining LLDPE resin and CaCO₃ powder using American Leistritz Extruder Corporation model ZSE 27 (Figure 2(a)). The extrusion equipment is equipped with a co-rotating intermeshing twin-screw extruder with numbers of heating zones. Schenck AccuRate gravimetric feeders were utilized to regulate the quantity of LLDPE accurately, and CaCO₃ supplied into the extruder. Then, the polymer strands enter a long water bath and cut to 3mm nominal length. Then, the pelletized composite resin was dried in an indirect heated dehumidifying oven (Figure 2(b)) at 60 °C for 7 hours. It was then kept in sealed moisture barrier bags before mixing with asphalt binder ^[26].

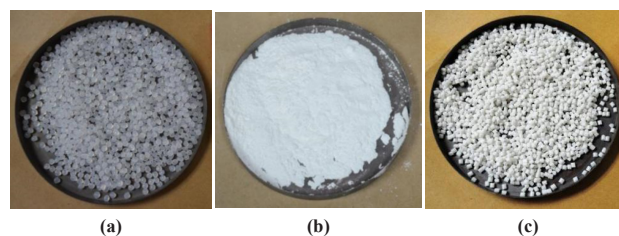


Figure 1. (a) LLDPE Pellets, (b) CaCO₃ Powder, (c) PECC Pellets

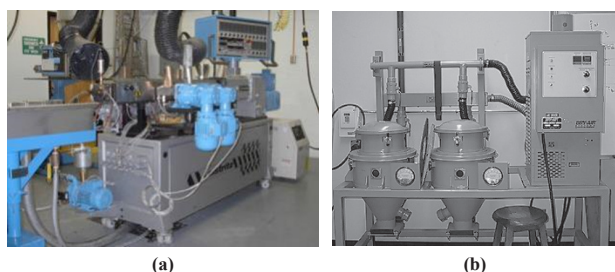


Figure 2. (a) 27mm Twin Screw American Leistritz Extruder, (b) Air Dryer dehumidifying oven

3. Specimen Preparation

3.1 Preparation of Modified Asphalt Binder

A high shear mixer was used for the preparation of modified asphalt binders. The shear mixer was used to ensure the modifiers are consistently dispersed in the asphalt binder at designated speed and time interval. The temperatures used to produce modified asphalt binders are shown in Table 2. During the production of modified asphalt binder, approximately 500g of asphalt binder PG58-28 was transferred into a one-litre metal can. Prior to the blending process of modifier with asphalt binder, the required quantity of modifier was added into the similar metal can that contain asphalt binder and was heated up in an oven at the designated temperature for two hours. Later, the asphalt binder and modifier were blended using a high shear mixer at the specified rotational speed of 5000 rpm for 45 minutes. In this study, due to great differences in density between LLDPE and CaCO_3 , the amounts of modifiers were controlled based on the volume of asphalt binder. The quantities of modifier incorporated in the asphalt binder are summarised in Table 2.

Table 2. Temperatures used for modified asphalt preparation

Sample Designations (% is based on volume)	Modifier	Production Temperature (°C)
3 vol% LLDPE	LLDPE pellets	175
6 vol% LLDPE		
3 vol% CaCO_3	CaCO_3 powder	160
6 vol% CaCO_3		
3 vol% PECC	PECC pellets	170
6 vol% PECC		

3.2 Preparations of Asphalt Mixture

The asphalt mixture was mixed using a bucket mixer. The loose mixture was then compacted using gyratory com-

pactor with 86 gyrations. In order to simulate the short-term ageing process that typically took place during the production of asphalt mixture, the loose asphalt mixture was conditioned in an oven for two hours. The compaction process takes place after the short-term simulation process. The Superpave specifications [27-29] were referred.

4. Asphalt Binder Behaviour

4.1 Volatile Loss

The quantification of asphalt binder volatiles lost (mass loss) during the short-term ageing process was conducted through the rolling thin film oven (RTFO) test. In this test, the light molecules from binder were driven off, increasing the asphalt's viscosity. The process was done by inducing the occurrence of heat and flowing air on a thin film of asphaltic material. Table 3 shows the mean mass loss test results. Most of the binders have low volatiles lost during the ageing process, where the values are less than one wt% of asphalt binder based on the Superpave Specification. The 6 vol% LLDPE sample has exhibited the highest volatile loss, which is about 0.116 wt% of asphalt binder. This indicates the modification process using LLDPE and CaCO_3 are not results in changes of the chemical structure of the asphalt binder. According to a study conducted by Fang et al. [14], the asphalt binder modified with a higher percentage of modifier has higher mean mass loss value, except the PECC's modified asphalt binder that presented the opposite trend.

Table 3. Mean mass loss values using RTFO aging process

Sample (% is based on volume of asphalt binder)	Results (Mean Mass Loss \pm Std Dev, n=3)
PG58-28	0.097 \pm 0.0010 wt%
3 vol% CaCO_3	0.095 \pm 0.0030 wt%
6 vol% CaCO_3	0.105 \pm 0.0005 wt%
3 vol% LLDPE	0.097 \pm 0.0005 wt%
6 vol% LLDPE	0.116 \pm 0.0040 wt%
3 vol% PECC	0.105 \pm 0.0020 wt%
6 vol% PECC	0.094 \pm 0.0030 wt%
*Requirement: Percent loss should be less than 1 wt%	

4.2 Rotational Viscosity

The rotational viscometer with #27 spindle was used to evaluate the viscosity of asphalt binders at service temperature. It is essential to ensure the asphalt binder

is pump-able and ease to handle prior to use in the mixture production stage, and in an ideal condition for the compaction in the field. This test examines the torque value needed to uphold a constant rotational speed (20 rpm) of a cylindrical spindle under a consistent temperature. The recorded torque value is then presented as viscosity.

The mean rotational viscosity test results are presented in Figures 3 to 5. The addition of modifier in asphalt binder results in higher viscosity compared to the neat binder (PG58-28). Higher percentages of modifiers have led to higher viscosity values. Adding LLDPE to asphalt binders has considerably increased the asphalt binders' viscosity and stiffness, which can be contributed to better resistance to rutting. The asphalt binder prepared with 6 vol% LLDPE indicates the highest viscosity compared to other modified asphalt binders. Whereas it has been translated into proportionally higher mixing and compaction temperature compared to PG 58-28 (Table 4). All samples are found to fulfil the Superpave requirement on rotational viscosity's value, whereas the RV readings should be lower than 3 Pa.s at 135 °C.

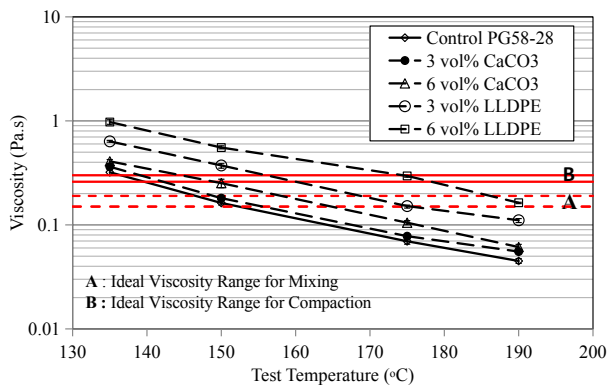


Figure 3. Effects of CaCO₃ and LLDPE modifiers on asphalt viscosity

Referring to Figure 3, addition of CaCO₃ into asphalt binders has escalated the binder viscosity, which is in a range of 13 – 23.1% and 27.3 – 55.1% for the asphalt binders modified with 3 vol% CaCO₃ and 6 vol% CaCO₃, respectively. A higher amount of CaCO₃ has led to a higher viscosity as a result of the presence of lime powder that increase the stiffness of binders. The presence of LLDPE molecules in asphalt binders has further increased the viscosity of the asphalt binders. The 3 vol% LLDPE's modified asphalt binder has doubled the neat asphalt binder's (PG 58-28) viscosity. Meanwhile, the modified asphalt binder prepared with 6 vol% LLDPE was found to have the highest viscosity compared to other modified asphalt binders, and three times greater compared to the neat asphalt binder.

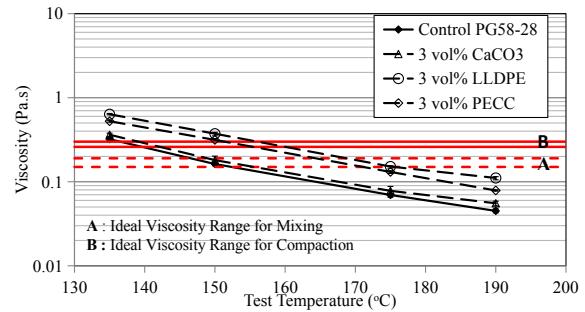


Figure 4. The viscosity of modified asphalt binders at 3 vol% of asphalt binder

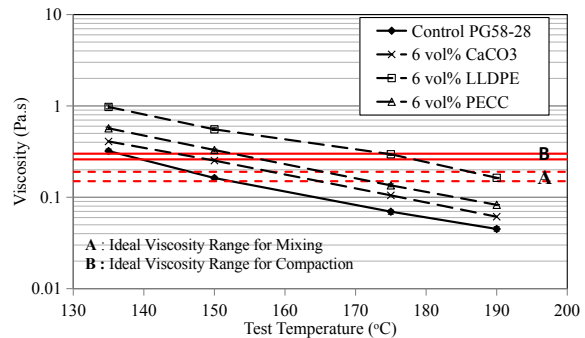


Figure 5. The viscosity of modified asphalt binders at 6 vol% of asphalt binder

Figures 4 and 5 illustrate the outcomes of asphalt binders modified using LLDPE, CaCO₃, and PECC modifiers. Modified asphalt binders prepared using a combination of LLDPE and CaCO₃ (PECC modifier) have lower viscosity value compared to LLDPE's modified asphalt binder. The presence of CaCO₃ in PECC's modified asphalt binder has tuned down its viscosity compared to LLDPE's modified asphalt binders. This result could be correlated to a research conducted by Zhou et al. [30].

The optimal mixing and compaction temperatures for an asphalt mixture as stated by the Asphalt Institute are at a viscosity of 0.17 ± 0.02 Pa.s and 0.28 ± 0.03 Pa.s, respectively. The ideal mixing and compaction temperatures were obtained, and the results are shown in Table 4.

Table 4. Mixing and compaction temperatures of asphalt mixture based on the ideal viscosity ranges

Sample (% is based on the volume of asphalt binder)	Temperature (°C), n=3	
	Mixing	Compaction
PG 58-28	150	140
3 vol% CaCO ₃	150	140
6 vol% CaCO ₃	160	150
3 vol% LLDPE	175	160
6 vol% LLDPE	185	175
3 vol% PECC	170	155
6 vol% PECC	170	155

4.3 Multiple Stress Creep Recovery

The dynamic shear rheometer test was conducted on asphalt binders at different ageing levels: un-ageing, short-term ageing and long-term ageing. The short-term ageing (STA) procedure is used to simulate ageing during mixing and construction processes. The STA ageing process was performed conforming to AASHTO T 240. While, the long-term ageing (LTA) was carried out conforming to AASHTO R 28 to simulate the ageing thru the pavement service life, approximately five to ten years period. The Multiple Stress Creep Recovery (MSCR) test was conducted using Bohlin CVO 120 high-resolution DSR.

The DSR with $G^*/\sin \delta$ (AASHTO M320) is the representative criterion for rutting projection of asphalt roadway. However, this technique has been appraised to deliver an improved estimation of the rutting performance of the modified asphalt binder, called multiple stress creep recovery (MSCR). This revised method computes the permanent strain concentrated in the binder after designated cycles of shear loading and unloading. In which, better resistance to rutting of the roadway are indicated by lower permanent shear strain value. In this study, the MSCR test was carried out by exposing the RTFO aged asphalt binder sample to repetitive creep and recovery process at elevated temperature. The test has been carried out in compliance with AASHTO TP 70-08 at 58 °C, which is the high-temperature grade of PG 58-28. The sample for MSCR testing was prepared in circular shaped asphalt binders with diameter of 25 mm and thickness of 1 mm. Two stress levels were assigned to the sample, which was 0.1 kPa and 3.2 kPa at one second loading time and nine seconds recovery time^[31,32]. The test was initiated with 0.1 kPa stress for ten cycles without time lags and continued with 3.2 kPa stress under the same number of cycles.

Non-recoverable compliance (J_{nr}) is the best method to substitute the existing Superpave method, $G^*/\sin \delta$ ($\omega = 10$ rad/s). Due to that, the resistance to rutting of asphalt binder is indicated by using non-recoverable compliance^[32,33]. Furthermore, in order to figure out the high-temperature viscoelastic deformation properties, the value of percentage recovery (R) was also determined^[32]. Where a better permanent deformation resistance is illustrated through lower J_{nr} value. Meanwhile, greater R-value specifies an improved rutting resistance.

Figure 6 shows the mean J_{nr} of specimens (n=3) tested under the 0.1 kPa and 3.2 kPa stresses respectively. Based on both figures, all the modified binders demonstrate lower mean J_{nr} values compared to control asphalt binder (PG 58-28), which directs a better resistance to rutting. Besides that, a higher percentage of modifiers

added to the control asphalt binders have resulted in better resistance to rutting. It was also found that the asphalt binder modified with 6 vol% LLDPE gave the lowest J_{nr} value compared to other asphalt binders, which indicates a better resistance to rutting. Theoretically, the lowest J_{nr} value indicates a better resistance to rutting under the respective stress.

Referring to Figure 6, CaCO_3 has successfully reduced the J_{nr} value of a neat asphalt binder at both stress levels, with higher amounts of CaCO_3 have resulted in lower J_{nr} values. The specimens prepared with 3 vol%, and 6 vol% CaCO_3 have better rutting resistance, which is about 9 % and 25 % higher compared to the control asphalt binder (PG 58-28). This result is consistent with previous studies which claimed the CaCO_3 could be used to reduce the susceptibility to rutting, due to an increase in stiffness^[34,35]. The CaCO_3 is typically incorporated into the asphalt mixture using a dry process, which enhances adhesive bonding between aggregates and asphalt binder. Based on the previous study conducted by Button and Epps^[36], asphalt mixture prepared using lime through a dry process has a better performance regarding resilient modulus, tensile strength, and Marshall stability compared to asphalt mixtures prepared using lime through a wet process.

Asphalt binder modified using 3 vol% LLDPE exhibits the best resistance to rutting. However, the combination of CaCO_3 and LLDPE, which was denoted as 3 vol% PECC has reduced the material response to loading or recovery. It is due to the application of CaCO_3 in the modifier has slightly reduced the J_{nr} value compared to the modified binder that was prepared using LLDPE. Whereas, the CaCO_3 particles had resulted in a polymer-island effect that enlarged the distance between LLDPE molecules, which undermined the interaction force or bonding between material interface molecules^[15,30]. The 6 vol% LLDPE modified asphalt binder was found to have an improved rutting resistance compared to the mixture prepared using other modifiers. Bahia et al.^[37] mentioned that permanent deformation or creep is a repeated mechanism developed under sinusoidal loading pulses. However, some of the deformation will be recovered due to elastic stored energy in the materials, which dissipated in damping and permanent flow. The damping energy is recoverable if given enough time, and also believed to be the principal mechanism to improve the resistance to rutting of asphalt material. However, based on Lu and Isacson^[38], although the thermoplastic modifiers have enhanced the stiffness and viscosity of asphalt binder, it does not sufficiently help regarding elastic behaviour, which is crucial to improving the elastic behaviour of the modified binder.

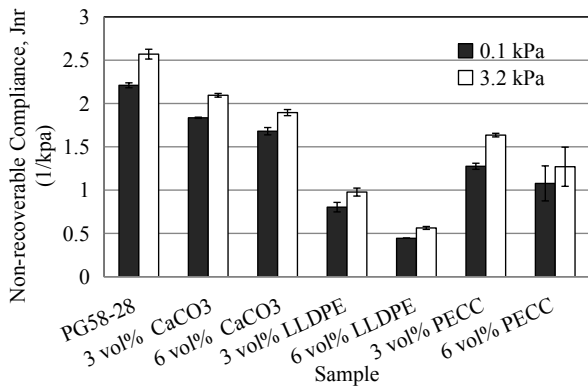


Figure 6. Results of MSCR at 0.1 kPa and 3.2 kPa

The mean percent recovery of the tested asphalt binders is presented in Figure 7. In general, the figure shows a positive trend of asphalt recovery with the increases of asphalt modifiers, excluding for the samples prepared using PECC modifier. The 6 vol% LLDPE has the highest percent recovery, which is about five times better compared to the control sample when tested at stress levels, 0.1 kPa and 3.2 kPa. It is also found that the percent recovery of 6 vol% CaCO₃ is comparable to 3 vol% CaCO₃ and control binder (PG58-28) at both stress levels.

Based on Figure 7, higher amounts of LLDPE have resulted in better R values that indicate a better resistance to rutting at both stress levels. The combination of LLDPE and CaCO₃ shows a negative effect on the recovery of asphalt binders. The highest reduction can be clearly spotted on the R-value results conducted on the 6 vol% PECC samples tested at 3.2kPa. This decrease is almost three times lower compared to specimen prepared using 6 vol% LLDPE.

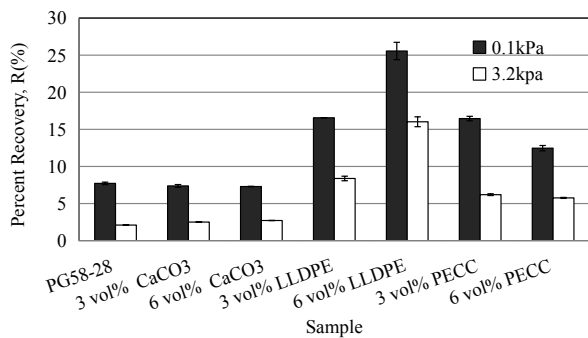


Figure 7. Results of percent recovery at 0.1 kPa and 3.2 kPa

4.4 Bending Beam Rheometer

The bending beam rheometer test was carried out to assess the low-temperature stiffness and relaxation properties of asphalt binders. It was assessed depending on the function of load and duration. An asphalt binder's ability to with-

stand low-temperature cracking was portrayed through this testing. The test was completed in accordance with AASHTO T 313. A simply supported beam of asphalt binder was subjected to a constant load of 980 mN for a duration of four minutes. Over time, the creep stiffness and m-value of the sample were documented at three low temperatures. The test was conducted at three different temperature of -12 °C, -18 °C and -24 °C to outline the critical cracking temperature of control and modified binders.

Figure 8 shows the low limiting temperature or T_{cr} for each binder. Generally, all the modified asphalt binders have revealed comparable performance on resistance to low-temperature cracking, except for the sample prepared using the LLDPE modifier. It can be deduced that integrating the LLDPE modifier had remarkably reduced the low-temperature grade of the asphalt binder. Whereas the thermal cracking might occur at -22 °C (3 vol% LLDPE) and -27.8°C (6 vol% LLDPE), compared to the control asphalt binder that could withstand the thermal cracking as low as -30.5 °C based on the BBR test.

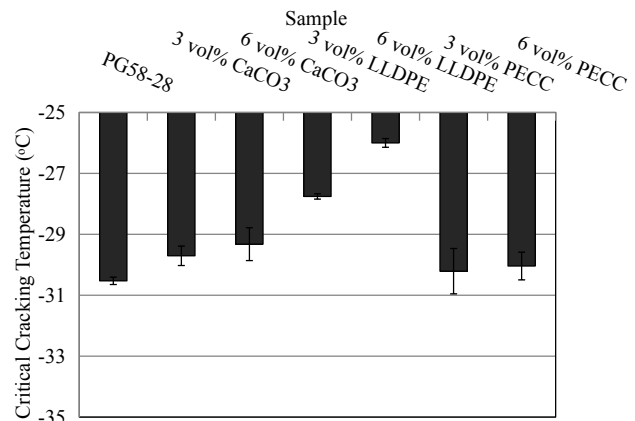


Figure 8. The critical cracking temperature of control binder and modified asphalt binder

Figures 9 (a) to (c) show the equipment used for the characterization of asphalt binders based on the viscosity, binder recovery, and the critical cracking temperature, respectively.

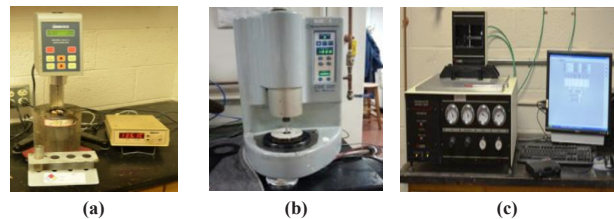


Figure 9. Equipment used for the binder test (a) Rotational viscometer, (b) Dynamic shear rheometer, (c) Bending beam rheometer

5. Asphalt Mixture Performance

5.1 Four-Point Beam Fatigue

The four-point beam fatigue test was utilized to evaluate the fatigue life of the asphalt mixture subjected to the repetitive bending till failure. The fatigue failure of the asphalt mixture was defined as a 50 % reduction of initial stiffness. In this test, a frequency of 10 Hz and 400 micro-strains (constant strain) was used for all samples. The test was conducted following AASHTO T321.

Figure 10 shows the mean values (n=3) from the four-point beam fatigue test. Based on the result, the specimen prepared with CaCO₃ is discovered to have the highest fatigue life compared to other samples, including the control sample. A higher proportion of CaCO₃ has led to better resistance to fatigue cracking. The fatigue life of the asphalt mixture prepared using 6 vol% CaCO₃ is 1.5 times greater compared to the control sample (PG58-28). Based on the previous studies, application of CaCO₃ also has successfully improved the ageing resistance, resistance to fracture adhesive bonding, and dynamic and residual stability which all contribute to the fatigue life of asphalt mixture [18,39]. However, the incorporation of LLDPE has weakened the fatigue life of asphalt mixtures. The lowest fatigue life value was found in 6 vol% LLDPE, which is 7.5 times lower compared to the control asphalt mixture. This result can be linked to an earlier study conducted by Lu and Isacson [38], whereas although the thermoplastic modifiers have enhanced the stiffness and viscosity of asphalt binder, it does not sufficiently help regarding elastic behaviour, which is crucial to improving the elastic behaviour of the modified binder.

Furthermore, the combination of LLDPE and CaCO₃ has slightly improved the fatigue life of asphalt mixture compared to specimen prepared using LLDPE. It is due to the appearance of CaCO₃ that has reduced the stiffness of the asphalt binder (based on the RV test results), which can be seen in Figures 4 and 5. However, higher amounts of PECC modifier incorporated into asphalt mixtures have resulted in lower resistance to fatigue cracking. The inconsistent results could be due to the inadequate time of mixing during the preparation of modified asphalt binder or asphalt mixture. It is essential to limit the amount of LLDPE modifier in the asphalt binder to ensure excellent resistance to fatigue cracking.

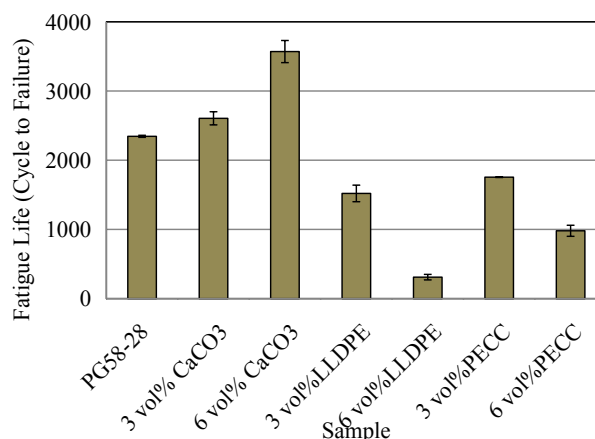


Figure 10. Comparison of fatigue life cycle values between CaCO₃, LLDPE and PECC

5.2 Moisture Damage

The tensile strength ratio (TSR) was used to assess the moisture susceptibility of asphalt mixture via tensile strength of specimens. Indirect tensile strength (ITS) of asphalt mixtures under two different condition, namely dry and wet conditions were compared in order to evaluate the moisture susceptibility as shown in Figure 11. In this evaluation, the test was done following AASHTO T283. The asphalt mixtures were tested at the room temperature with constant loading speed at the rate of 0.085 mm/s. The asphalt mixtures were subjected to compression loads which act parallel to the vertical diameter plane. From Figure 11, only one type of trend can be observed where the addition of modifier improved the strength of asphalt mixture under both dry and wet condition compared to control specimens. However, PECC modified asphalt mixture tensile strength is much lower compared to LLDPE and CaCO₃ modified asphalt mixture.

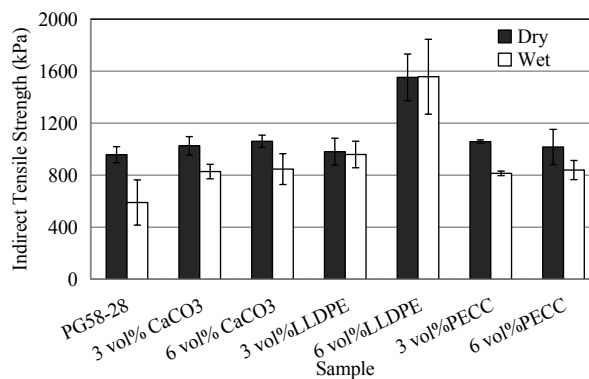


Figure 11. Comparison of ITS values between CaCO₃, LLDPE and PECC

Figure 12 shows the mean TSR results ($n=3$) for the control and modified asphalt mixtures prepared at two different percentages of each modifier. In general, most asphalt mixtures' TSR values are higher than 0.8, except the specimen prepared with 3 vol% PECC, which approximately 0.770. In this study, the CaCO_3 powder has been incorporated into the asphalt mixture using a wet process to ensure a similar preparation protocol for all samples. Through the wet process, the CaCO_3 powder (also other modifiers) was formerly blended with the asphalt binder using a high shear mixture before blending with the aggregates. This process is slightly different compared to the dry process, which is the typical approach of adding CaCO_3 into the asphalt mix. In the dry process, the CaCO_3 powder directly combines with the aggregates instead of blending it with asphalt binder. It is used to allow direct contact between CaCO_3 particles with aggregates and asphalt binder to improve the adhesive bonding in the mixture, hence better resistance to moisture damage. According to the result, the TSR values of 3 vol% CaCO_3 and 6 vol% CaCO_3 are not much different compared to the control sample because the CaCO_3 particles have been completely coated by the asphalt binder and not directly reacted with the aggregate surface.

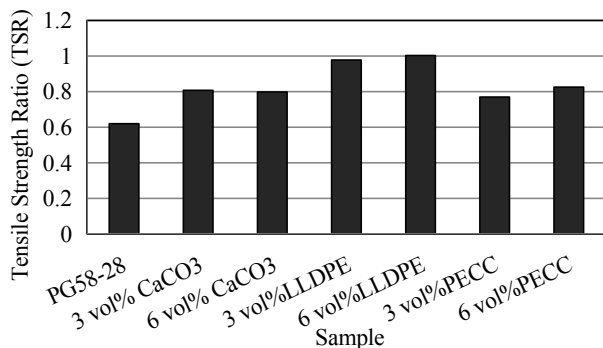


Figure 12. Mean tensile strength ratio results

Moreover, the incorporation of LLDPE has increased the TSR of asphalt mixtures as presented in Figure 12. This is particularly the case for the specimen prepared with 6 vol% LLDPE which has the highest ITS values in both conditions, dry and wet, as shown in Figure 11. This indicates the presence of LLDPE in asphalt mixture has altered the bonding between materials and prevents the HMA failure due to moisture damage through the freeze-thaw cycle. However, application of PECC modifier (a combination of LLDPE and CaCO_3) greatly undermined the resistance to moisture damage of asphalt mixtures with TSR values less than 0.8, and lower ITS values compared to specimens prepared with either LLDPE and CaCO_3 separately (Figure 11). The possible explanations

of this condition have been cited from Zhou et al. [30], where incorporation of CaCO_3 into polyethylene (PE) has reduced the specimen tensile strength. Zhou et al. further mentioned that CaCO_3 particles had enlarged the distance between LLDPE molecules, which undermined the interaction force or bonding between material interface molecules [30].

5.3 Dynamic Modulus

The dynamic modulus test (Figure 13) was done using UTM 100 from IPC as stated in AASHTO TP62-03 at different temperatures of $-10\text{ }^\circ\text{C}$, $4\text{ }^\circ\text{C}$, $21.3\text{ }^\circ\text{C}$ and $39.4\text{ }^\circ\text{C}$. Different frequencies ranging from 0.1 Hz to 25 Hz was used to conduct this test. The recoverable axial micro-strain was controlled within 75, and 125 microstrains to confirm the material was in a viscoelastic spectrum [40, 41]. This test is a response established under sinusoidal loading conditions, and the asphalt mixture sample is loaded under the compressive test. The applied stress and the subsequent recoverable axial strain response of the asphalt mixture sample have been measured continuously. The software automatically computed the dynamic modulus and phase angle of the asphalt mixture sample. The dynamic modulus is defined as the ratio of the amplitude stress (σ) and amplitude of the sinusoidal strain (ϵ) that results in a steady-state response at the same time and frequency.

Master curve technique had been used to analyze and compare the dynamic modulus (E^*) test results. The technique that was used to shift all E^* values at various frequencies and temperatures into one single curve is known as the sigmoidal fitting model. Through this method, the numbers of curves have been associated to form a single master curve which is based on a reference temperature of $-10\text{ }^\circ\text{C}$.

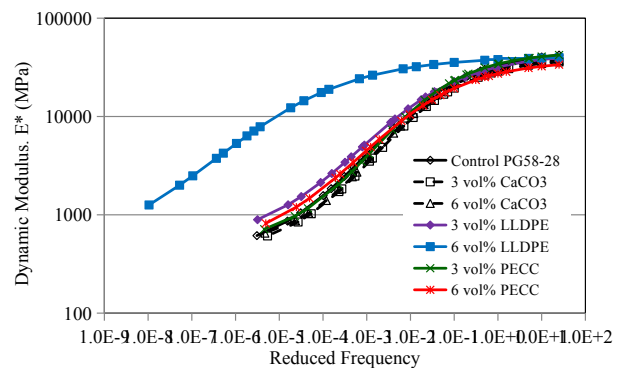


Figure 13. Mean dynamic modulus results of control and modified asphalt mixtures

6. Conclusions

Based on the results and analysis of this research, the conclusions can be drawn as follows:

(1) All modified asphalt binders have a low volatile loss during the short-term ageing process (less than 1 wt%). This indicates the modification process using LLDPE, CaCO₃, and their combination does not result in the changes of the chemical structure and chemical reaction in the asphalt binder. Referring to MSCR test results, the modified asphalt binders have better performance regarding high-temperature behaviour, at least 15 % higher compared to control binder.

(2) The viscosity of asphalt binder increases by incorporating the modifier, which significantly improved the mixing and compaction temperature compared to control asphalt binder. The highest viscosity values are produced by the modified asphalt binder prepared using 6 vol% LLDPE.

(3) Incorporation of 6 vol% LLDPE has remarkably reduced the low-temperature grade of the asphalt binder, where the thermal cracking could happen at -26 °C, compared to control asphalt binder that may withstand the thermal cracking at -30.5 °C based on the bending beam rheological testing results.

(4) In general, the results indicate that a higher percentage of modifier content has reduced the mixture air voids, hence lowered the optimum binder content required for the sample preparation. The decreases in air voids with increases of modifier content may be attributed to the modifier particles filling the voids.

(5) The decrease in fatigue life can be observed based on the four-point beam fatigue test where asphalt mixture prepared using LLDPE-related modifiers has the lowest fatigue life. However, the specimen prepared with CaCO₃ was discovered to have the highest fatigue life compared to control and other modified samples. CaCO₃ has significantly improved the fatigue performance of asphaltic mixtures, which were higher by about 11.1 % and 52.2 % for the specimen prepared using 3 vol% CaCO₃ and 6 vol% CaCO₃, respectively compared to control mixture.

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