



Utilization of Micronized Polyethylene Terephthalate (MPET) as a Filler in Asphalt Pavement

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Abstract: Nowadays, plastic pollution is one of the most critical issues related to sustainability development that need to be solved. Studies indicated that addition of certain amount of recycled plastic from Polyethylene Terephthalate (PET) in asphalt mixture has improved the engineering properties such as marshall stability, moisture resistance, rutting and fatigue resistance. However, the main challenge of direct incorporation of recycled plastic in binder is the thermal incompatibility of two phases which are glass transition and melting temperature. Previous research findings indicated that reduction in particles diameter to micro scale would decrease the PET melting point. Hence, this study investigates the potential of micronized PET from recycled plastic bottle as filler in asphalt pavement. Micronization process at laboratory was carried out to determine the appropriate method prior incorporating it into asphalt mixture. The micronized size of PET incorporated at 0%, 4%, 8%, and 12% by weight of filler is then evaluated for its volumetric properties, mechanical, and adhesion properties. The results showed that the volumetric properties of the asphalt were affected by the presence of micronized PET. The mechanical properties in terms of Resilient Modulus and Indirect Tensile Strength showed increment with addition of micronized PET. Similar trend also observed on adhesion properties. Hence, the findings indicated the potential of utilizing the recycled MPET for local road construction.

Keywords: Micronized, polyethylene terephthalate, filler, volumetric, mechanical

1. Introduction

In literature review, plastic manufacturing's main application contributed from 39.9% of packaging industry, 19.8% of construction industry, 9.9% of automation industry, and 6.2% of electronics (Zair, et al., 2021). Furthermore, approximately 30% of total plastic waste material is processed by plastic recycling plant, while 70% is disposed through landfills that brings about changes in ecological system (Haider, et al., 2019). Due to their nonbiodegradable nature, plastic waste takes as long as five hundred years to fully decompose, and directly leads to negative environmental impact, through water bodies pollution, aquatics lives extinction, addition of carbon emission, and cities' aesthetic damage (Dalhat, et al., 2019). The best plastic management strategy is identified as recycling, given its high recyclability potential,

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inexpensive and easily mould into solid objects. However, it has been a global challenge to execute the appropriate disposal of waste plastics as the demand of manufacturing of various forms of plastics continuously produced.

Alternatively, waste plastics are incorporated with asphalt mixtures as modifiers to enhance its performance properties as well as to gain economic and environmental benefits. Polyethylene Terephthalate (PET) is of thermoplastics polyester material, which globally used in the food and beverage industry to produce plastic products such as water bottles, disposable dishes, and plastic containers. Several studies have been conducted in incorporating various forms of waste plastics through several approaches into asphalt mixtures to enhance modified mixtures' performance properties (Xu, et al., 2022). Researchers have also produced various particle sizes of waste PET as modifiers through uncomplicated method such as cutting, shredding, and milling. For instance, Moghaddam et al. (2012) and Ahmadinia et al. (2012) cut waste PET into small parts, then crushed by using crushing machine before sieved to pass 2.36mm and 1.18mm particle size, respectively. Ghabchi et al. (2021) cut waste PET into small pieces before milled by milling device to pass sieve size ranged from 0.037mm to 0.075mm. While studies indicate potential of incorporating various size of waste PET in asphalt mixes, their inconsistent sizes also implied the effect of PET particle size on the properties of modified mixture.

According to Ghabchi et al. (2021), large particle size in PET does not provide significant improvement on asphalt binder performance when it is used as aggregate replacement. Hence, the main mechanical properties of PET as asphalt modifiers could not be fully utilized. Other than that, the production of crushed PET as aggregate replacement is found to be more costly than aggregate production in the quarry. There are two major factors that affecting PET's physical and mechanical properties which are crystallization and orientation (Silva, et al., 2018). Polymer chains are rigid below glass transition temperature, after it reached the glass transition temperature, the chains are crystallized, more flexible and can be disentangled under stress. Considering the high melting (260 °C) and glass transition temperature (69 °C) of PET, it has become a challenge to directly incorporate PET in asphalt binder under mixing and compaction temperature of 165 °C and 125 °C, respectively. The thermal incompatibility of these two phases hindered PET to fully melt, as it only provides rubbery behaviour and a soft structure in mixing and compacting temperature. One possible alternative to further soften PET and partially melt it at lower melting temperature is by reducing its particle dimension to superfine sizes [7,8]. Ghabchi et al. (2021) found that the incorporation of MPET in asphalt mixes shown better mechanical properties as it exhibited higher cracking and moisture damage resistance. The MPET is micronized via drying and freezing process before milled into milling devices. Similar micronization process also applied by a study done by Silva et al. (2018), that investigated mechanical performance of MPET-modified asphalt binder. It was suggested that MPET improved asphalt mixtures' mechanical behaviour through the increasing parameter of indirect tensile strength, resilient modulus, fatigue life, and moisture damage resistance. Ferreira et al. (2022) crushed PET via different blades and milled until it micronized to be utilized as natural sand replacement in asphalt mixture. It was found that adding MPET in the mixture significantly contribute to resistance to moisture damage through asphalt binder-aggregate adhesion improvement. The volumetric properties of increasing MPET content also investigated where binder film thickness decreased, maximum specific gravity reduced, and volume of voids increased.

The abovementioned studies indicated the effect of different MPET micronization process on the mechanical and volumetric properties of asphalt mixes. Thus, this research was conducted to investigate the effects of MPET on volumetric, mechanical and adhesion properties of asphalt mixtures. The outcome of this study is expected to introduce different MPET micronization approach as plastic recycling alternative method to reduce increasing plastic waste material production, and simultaneously contribute to sustainable practice in infrastructure construction work.

2. Materials and Mix Design

2.1 Filler

Filler is used for the design mix for both controlled sample and MPET incorporation as filler replacement. In this study, quarry dust is used as filler as it is available from quarries compared to other fillers that will incur transportation cost from factories. The filler is supplied by Hap Seng Papar Quarry Sdn. Bhd. and sieved to pass 0.075mm size.

2.2 Aggregate

The aggregate used in this study were also obtained from Hap Seng Papar Quarry Sdn. Bhd., Sabah. The gradation of aggregate for hot mix asphalt AC14 was proposed in accordance to gradation limit of hot mix asphalt (HMA) based on Public Work Department (PWD), Malaysia (JKR/SPJ/2008-S4, 2008). Table 1 showed the physical properties of aggregate.

Table 1 - Physical properties of aggregate

Property	Value	Standard test method
Flakiness index (%)	13.78	BS 812, Part 3
Aggregate crushing value	23.47	BS 812 Part 110 – 90
Aggregate impact value (%)	10.59	BS 812 Part 3, 1975
Los Angeles abrasion (%)	23.3	AASHTO T 96

Water Absorption – coarser than 10 mm (%)	1.11	ASTM D 570
Water Absorption – smaller than 9.5 mm (%)	1.56	ASTM D 570
Specific gravity test (coarse)	2.56	AASHTO T 104 - 771
Soundness (coarse) (%)	0.94	AASHTO T 104 - 771
Soundness (fine) (%)	3.33	AASHTO T 104 - 771

2.3 Asphalt Binder

Bitumen penetration grade of 60/70 was used in this study as it is conventionally used in road construction and maintenance in Malaysia. Table 2 below presents the bitumen properties that passed the minimum properties standard requirement in industrial practice of PWD (JKR/SPJ/2008-S4, 2008).

Table 2 - Properties of bitumen

Property	Value	Standard test method
Ductility at 25 °C (cm)	127.3	ASTM D 113
Penetration at 25 °C (0.1 mm)	68	ASTM D 5
Softening Point (°C)	54	ASTM D 36

2.4 Micronized PET

PET is the most common thermoplastic polymer which consists of $C_{10}H_8O_4$ monomer units that belongs to polyester family. PET has density of $1.38g/cm^3$ at 20 °C, melting point of 260 °C, and glass transition temperature of 69 °C (Callister, 2020). In this study, PET underwent micronization process to be utilized as filler replacement in asphalt mixture. In this process, water bottles were collected from consumers' water bottles, and had its labels and caps removed. The water bottles were then cut to smaller pieces and dried at 45 °C to remove all moisture. They were heated in the oven at 3 different temperatures which are 70 °C, 150 °C, and 190 °C for 2 hours before being crushed using Los Angeles (LA) abrasion machine. Lastly, crushed PET was sieved to obtain single size PET particle that passed through 0.075mm size.

3. Experimental Design

3.1 Design Mix

Design mix is essential to determine the Optimum Bitumen Content (OBC). In this study, MPET was added as filler replacement. Specimens were prepared with bitumen content ranged between 4% to 6% with 0.5% increment. Then, MPET was added to the aggregate at 4%, 8%, and 12% by weight of filler. Mixtures were mixed and compacted at 160 °C and 150 °C respectively which is based on premix plant requirement. Volumetric and strength properties of each sample were analysed. Volumetric properties are percentage of voids in total mixture (VTM), voids in aggregate filled with bitumen (VFB) and bulk specific gravity of mixture (Gmb) (Razzaq, et al., 2018). Based on the analysis, OBC was proposed. The standard was based on PWD and shown on Table 3.

Table 3 - Specification parameters for OBC of AC14

Parameters	Wearing course (AC14)
Stability, S	>8000 N
Flow, F	2.0 – 4.0 mm
Stiffness, S/F	>2000 N/mm
Voids in Total Mix (VTM)	3.0 – 5.0 %
Voids in aggregate filled with bitumen (VFB)	70 – 80%

3.2 Resilience Modulus Test

The resilient modulus test is performed in accordance to ASTM D4123. The test was carried out at temperatures of 25 °C by repeatedly applying compressive loads to the specimen in a haversine waveform and prior to research, the specimens were conditioned for 4 hours \pm 5 minutes at 25 °C. With a pulse width of 0.1 second, a haversine load of 1kN peak force was applied along with a rest time of 0.9 second with an assumed Poisson ratio of 0.40 (Omar, et al., 2018). The compressive load was applied along the curved plane of an asphalt mixture cylindrical sample. The final readings of

resilience modulus were taken according to the average values of the 3 samples for every MPET content in asphalt mixtures.

3.3 Indirect Tensile Strength Test

The Indirect Tensile Strength Test (ITS) was performed according to UNI EN 12697-23. The height of the specimen was determined in accordance with ASTM D3549. The specimen were conditioned for 4 hours at temperature of 25 °C in the chamber. The vertical compressive ramp load was applied until the maximum load was reached. Equation (1) shows the formula used to calculate the ITS (Omar, et al., 2018).

$$\text{Indirect Tensile Strength} = (2000 \times P) / (\pi \times t \times D) \quad (1)$$

Where:

P = Maximum load, N

t = Specimen Height, mm

D = Specimen diameter, mm

3.4 Adhesion Test

The boiling test procedure was based on ASTM D3625-96. The asphalt mixtures were prepared by adopting the plant requirement. Asphalt binders and aggregates were stored in the oven at mixing temperature for 2 hours and then mixed using a mixer. After mixing, the loose mix was cured in the oven for the prescribed time. Then, the loose mixtures were boiled for 15 - 20 minutes. Visual inspection of specimen post boiling was carried out following the UNI EN 12697-11.

4. Result and Discussion

4.1 Micronization of PET as Fillers

PET has melting temperature of 260 °C, and it experienced thermal incompatibility when added to the mixture as it does not melt at asphalt mixture’s melting point temperature. Therefore, reduction in particles diameter to micro scale bring about reduction to PET melting point, which is a phenomenon known as melting point depression (Ghabchi, et al., 2021). This study applied heat energy to increase the efficiency of PET crushing process through LA abrasion machine. Based on the result in Figure 1, it was found that temperature of 190 °C was the most optimum temperature compared to its melting point of 260 °C. The other two temperature could not produce smaller particle size that could pass sieve size of 0.075mm. In view of PET micronization, Silva et. al. (2018) stated that reduced particle size of PET is obtained through a traditional friction-based micronization technique. Therefore, as MPET is heated to 72 °C, crystallization slowly occurred, and it started to become opaque, stiffer, and less flexible. Zair et. al. (2021) also suggested that reducing the particle dimensions of PET to superfine sizes will further soften PET in liquid asphalt and partially melt at lower temperature than its melting point. Based on this finding, it showed that heating PET at a temperature of 190 °C for 2 hours prior to crushing process using LA abrasion machine is applicable to produce MPET and incorporate it with asphalt mixture as filler replacement. Apart from that, the energy consumption for PET to reach its melting point could be reduced as lower heating temperature of PET is applied before crushing process which in turn reduced the cost of heating the PET especially when it comes to its usage as asphalt modifier.

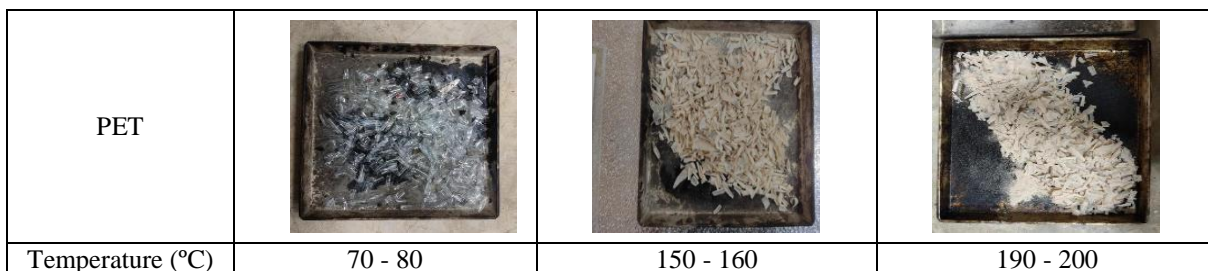


Fig.1 - Physical appearance of PET after being heated

4.2 Optimum Binder Content Determination

Based on the specifications for OBC of HMA for AC14, the required volumetric data and resilient modulus of the HMA containing PET were analysed. Table 4 presents the OBC of various MPET content. The OBC for control sample (0% MPET) is similar with all MPET content which indicated that OBC was not affected by incorporation of MPET as filler replacement to asphalt structure.

Table 4 - OBC for all samples

MPET Percentages (%)	Bitumen Content (%)
0	5.2
4	5.2
8	5.2
12	5.2

4.3 Volumetric Properties

Table 5 presents the volumetric properties which are Voids in Total Mixture (VTM), Void Filled with Bitumen (VFB) and bulk specific density (Gmb). In general, the addition of MPET affected the volumetric properties.

Table 5 - Volumetric properties of modified asphalt

MPET Percentages (%)	Bulk Specific Gravity, Gmb (kg/m ³)	Voids filled Bitumen, VFB (%)	Voids in total mixture, VTM (%)
0	2.354	72.748	3.394
4	2.357	73.310	3.273
8	2.361	74.019	3.122
12	2.362	74.197	3.085

4.4 Density

The densities of asphalt mixtures at different percentage of MPET are shown in figure 2. The density of asphalt mixture increases as the percentage of MPET increases. The MPET-modified asphalt mixture with 12% MPET produced the highest density which is 2.36kg/m³.

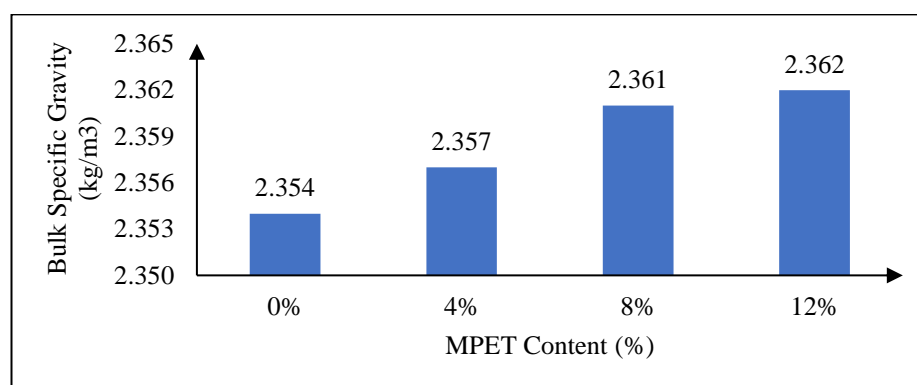


Fig. 2 - Density of asphalt mixture containing different MPET percentage

4.5 Voids in Total Mixture

Based on the result presented in figure 3, the VTM decreased with MPET percentages. Air void is closely tied to how the mixture is compacted. Hence, as the small particle were filled with aggregate during compaction, air void was diminished. The result indicates that presence of MPET had lowered air voids in asphalt mixture. Based on PWD specification, 3% to 5 % of air void is required to account for the increased compaction brought on by the traffic load (JKR/SPJ/2008-S4, 2008). The addition of highest MPET content of 12% has air voids of 3.09%, which is still in the range. Therefore, addition of MPET content more than 12% would result in lower air void and may cause bleeding of pavement.

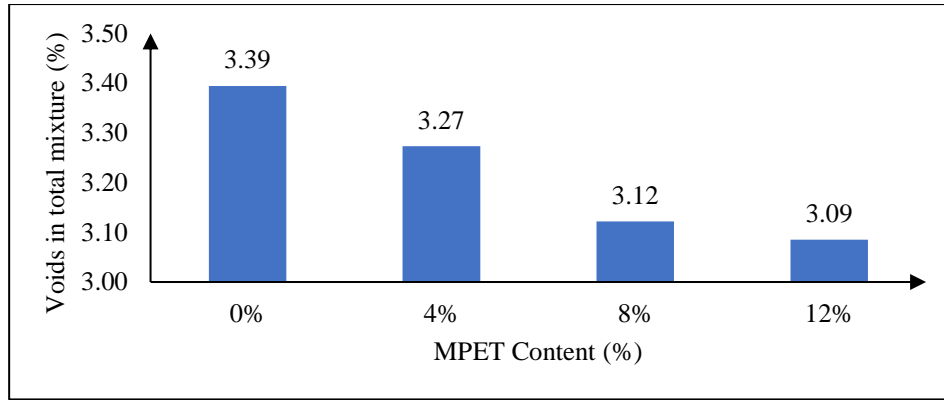


Fig. 3 - VTM of asphalt mixture at different MPET percentage

4.6 Voids Filled with Bitumen

Figure 4 exhibit the voids filled with bitumen against addition of MPET percentage. The result showed were similar with density where total air voids between aggregates filled with asphalt increased with the addition of MPET. PWD specification stated that VFB specimen value should range between 70% to 80% (JKR/SPJ/2008-S4, 2008). It could be seen the addition of MPET had increased the VFB yet still within range of PWD specification.

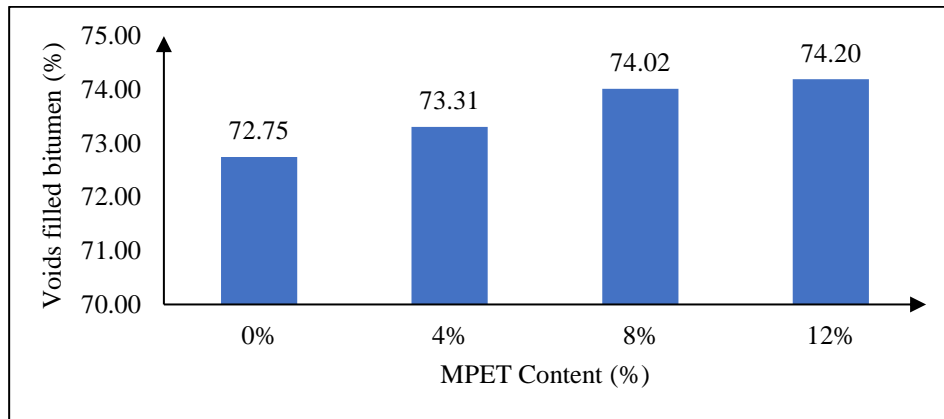


Fig. 4 -VFB of asphalt mixture at different MPET percentage

4.7 Mechanical Properties

The effect of MPET incorporation by weight of filler in asphalt mixture on mechanical properties was evaluated via Resilient Modulus and Indirect Tensile Strength tests.

4.7.1 Resilient Modulus Test

The resilient modulus test is conducted to determine the stiffness of the samples. Figure 5 shows the resilience modulus of asphalt mixture containing various MPET content. This shows that the addition of MPET content can improve the strength of the asphalt up to 12% replacement of fillers. The highest values obtain in this study is 4097.8 N/mm which is the incorporation of MPET for a value of 12% as filler replacement. The results show increment of stiffness in low temperatures which makes them susceptible to cracking problems. This result also supported the study done by Silva et. al (2018) that has investigated the resilient modulus of MPET-modified asphalt binder as 25 °C and 40 °C. The resilient modulus produced better performance at 25 °C, and permanent deformation decreased when MPET is added to the mixture. Other than that, based on a study done by Dalhat et. al (2019), resilient modulus of recycled plastic waste with more fine sizes (sieve no. 8 to no.40) produced better moisture sensitivity with better resilient modulus results. Previous study gives an overview that smaller particle size of PET lead to a better strength performance in term of resilient modulus, resulting better resistance to cracking and moisture damage.

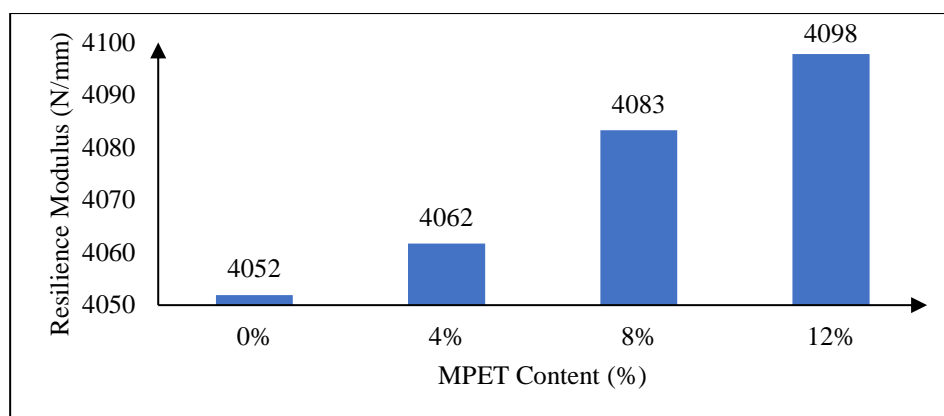


Fig. 5 - Resilience Modulus of asphalt mixture at different MPET percentage

4.7.2 Indirect Tensile Strength Test

This test was done to determine tensile strength of asphalt mixture and investigate its rutting and cracking properties. Based on the results shown in figure 6, the strength properties of the asphalt improved from 480kPa to 542kPa with the addition of MPET content from 0% to 12% as filler replacement. This observation indicated that MPET-modified asphalt mixture could improve its resistance to rutting. This improvement was also proven by study done by Ghabchi et. al. that found better resistance to rutting and raveling through investigation of PG 58-28 asphalt incorporation with 5%, 10%, 15%, and 20% of MPET into the mixture for both dry and moisture conditioned samples (Ghabchi, et al., 2021). In the study, both dry and moisture conditioned samples showed improvement on tensile strength which are 34% higher on dry condition incorporating 20% of MPET, and 50% higher on moisture conditioned incorporating 20% MPET compared to 0% of MPET content in asphalt mixture. Furthermore, a study done by Silva et. al. that investigated mechanical properties on MPET-modified asphalt mixture, also concluded that the presence of MPET has increased the parameter of indirect tensile strength and other strength parameters (Silva, et al., 2018).

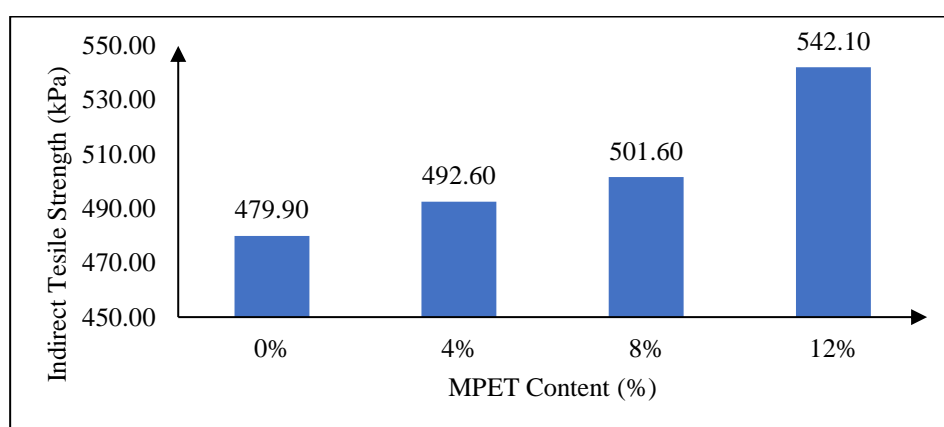


Fig. 6 - Indirect tensile strength of asphalt mixture at different MPET percentage

4.8 Adhesion Test

Figure 7 shows the visual observation on loose mixture after water boiling test to check the adhesion properties. Based on visual observation, it was found that the coating between aggregate and asphalt is more even as content of MPET is increased. When aggregates are coated uniformly, it enables better bonding between aggregates. The 12% MPET content showed a much darker color and more even coating distribution compared to 0%, 4%, and 8% of MPET content. These results indicated that higher percentage of MPET can increase the adhesion of the asphalt and thus resulting to better resistance to moisture damage. According to Zair et. al. (2021), MPET had least impact on moisture damage in asphalt mixture likely due to the reason that PET particles improved the mastic cohesion via mechanical adhesion, surface energy, and binder rheology. Furthermore, results from Binder Bond Strength and Tensile Strength Ratio test and Surface Free Energy analysis from study done by Ghabchi (2021), also obtained similar result where there is an improved adhesion and moisture damage resistance when incorporating MPET in asphalt mixtures with different types of aggregates.

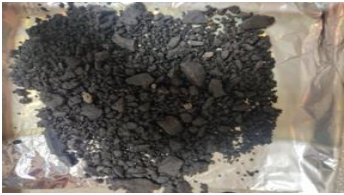











MPET Content (%)	Samples		
	Sample 1	Sample 2	Sample 3
0%			
4%			
8%			
12%			

Fig. 7 -Water boiling test results

5. Conclusion

Based on the result and analysis, it can be concluded that the addition of MPET as filler replacement affected the volumetric properties as well as mechanical properties of HMA. The addition of MPET caused VTM to decrease yet increased its density and VFB. Furthermore, the method of micronizing PET through heating at 190 °C before crushing has lowered the melting temperature of PET (260 °C) and is proven to be applicable in producing MPET-modified asphalt mixture with improved performance. Other than that, the presence of MPET did not affect the OBC.

The mechanical properties of the asphalt increased with the addition of MPET as filler replacement. The increasing values of Resilience Modulus and Indirect Tensile Strength through incorporation of higher amount of MPET in asphalt mixtures indicated higher resistance to rutting and permanent deformation. An improvement in the adhesion properties from MPET-modified asphalt mixture compared to conventional mixture is also observed. The presence of MPET indicated that aggregates are having better coating compared to asphalt mixture containing 0% of MPET.

The best replacement percentage of MPET addition as filler that can effectively improve asphalt performance is at 12%. Lastly, it would be beneficial to incorporate MPET with asphalt pavement as not only improvement on asphalt performance is achieved, but also serves as an environmentally sound method to maintain sustainability that has becoming our major concerns nowadays.

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