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Asphalt Binder Modification with High-Density Polyethylene Polymer and Low-Density Polyethylene Polymer – Efficiency of Conducting Semi-Wet Mixing Process

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ABSTRACT

In this research, the performance of asphalt mixtures modified with polyethylene polymer (PE) by adding 2%, 4%, and 6% percentages was evaluated. Two kinds of PE are employed: Low-Density PE (LDPE) and High-Density PE (HDPE). The semi-wet mixing technique (SWM) was conducted to avoid stability issue for PE-modified binder during storage condition. Many experimental tests were conducted to evaluate the ability of these mixtures to withstand the effects of loads and moisture. The hardness index of these mixtures was also measured to determine their resistance to the effects of high temperatures without causing permanent deformations. The results showed that adding PE led to a remarkable enhancement in the performance of PE-modified mixtures. The improvement in stability reached 113.36% when using HDPE and it was 86.19% for LDPE. For moisture resistance, it improved by 10.42% and 9.91% when using HDPE and LDPE, respectively. The modified mixtures also showed higher hardness index compared to the standard mixture. According to the outcomes of this research, it can be concluded that the optimum percentage for using PE polymer as a modifier in bitumen is 6% and HDPE is more effective than LDPE. Overall, the SWM is easier and more economical compared to the wet mixing.

Keywords: HDPE, LDPE, semi-wet mixing, Marshall properties, moisture damage.

INTRODUCTION

The roads are deteriorating rapidly due to a combination of increased traffic, harsh environmental condition, and insufficient maintenance caused by a shortage of funds. Avoiding this deterioration has always been a hard challenge for road pavement engineers and authorities (Abd and Latief, 2024). In addition, the development of the road network has become an urgent necessity for many vital reasons, including increasing traffic, economic development, and urban expansion. Thus, several measures must be taken to extend the service life of pavements and to provide a better ride quality (Eskandarsefat et al., 2022; Latief, 2019). Among the strategies applied to tackle these challenges, the utilization of polymer-modified asphalt binders emerges as a viable solution to meet contemporary pavement performance standards and has proven to be an effective approach. Over the years, this approach is perceived as both rational and practical, offering economic benefits in tandem with enhanced pavement performance (AL-Azawee and Latief, 2020).

There is an urgent need to modify the asphalt binder using synthetic polymers such as rubber and plastic to enhance the performance of bitumen and boost its resistance to various environmental factors (Mohammed et al., 2024). Incorporating polymer into the asphalt mixture enhances its flexibility at high and intermediate temperatures, hence enhancing its resistance to rutting and fatigue. Polymers typically have a higher viscosity than asphalt binders and exhibit enhanced adhesion to aggregate. This leads to the formation of thicker binder coatings, which will take longer to become brittle due to oxidation, hence improving the longevity of the mixes (Asmael and Waheed, 2018).

PE is widely regarded as the most prevalent type of plastic globally. The PE polymer has a relatively crystalline structure. It possesses exceptional resistance to chemicals, as well as favourable qualities in terms of fatigue and wear resistance. Additionally, it exhibits a diverse variety of features. The structure is fairly simplistic. PE is a polymer composed of a series of carbon atoms linked together in a chain, with two hydrogen atoms bonded to each carbon atom. These objects have a low weight and offer excellent resistance to organic solvents, along with low rates of moisture absorption (Awwad and Shbeeb, 2007). PE polymer can be mainly categorized into two groups; LDPE and HDPE.

The PE polymer functions as an aggregate replacement, binder modifier, mixture modifier, or a combination of these. There are two primary methods for including PE in hot asphalt mixture: the dry and wet processes. The dry process involves adding PE directly to hot aggregate and blended them at a specific temperature then added the heated asphalt binder to the PE-aggregate mix. Thin film of melted PE is formed to coat the aggregates surface. Meanwhile, the wet process involves adding PE to the binder to produce PE-modified binder (PEMB) which stored at certain conditions and can be used later in pavement production. In this method, blending of asphalt binder with PE polymer at high temperatures is a physicochemical process, in which PE absorbs the lightweight elements in binder to form a dispersion of viscoelastic phase (Chiang et al., 2024; Ghani et al., 2022). The dispersion of PE polymer in the mixture is reliant on factors like blending temperature, PE content, particle size, and mixing process. From economical perspective, using PE polymer in production of modified asphalt pavements can result in saving costs because this reduces the need for aggregate and virgin bitumen.

According to the researchers, wet process is currently more effective than dry process in enhance the performance of PE-modified asphalt pavement. Modifying the asphalt binder with LDPE and HDPE will improve the ability of this binder to withstand repeated loads and to provide better resistance to cracking (Mainieri et al., 2024; Lubis et al., 2020; Saadeh et al., 2024). Also, at low and high temperatures, the rheological properties of PEMB are enhanced (da Silva et al., 2024; Muhmood and Yahiya, 2024). In addition, these polymers have the ability to improve the resistance against rutting distress and reduce the effect of moisture damage of modified asphalt pavement (Abduljabbar et al., 2022; Elnaml et al., 2023; Jamil et al., 2023; Moussa et al., 2020). Nevertheless, wet process is more complex, excessive environmental impact, and highly cost than dry process since wet process requires several equipment and steps, long blending time, higher energy, and usage of solvent.

Despite the advantages of the wet process, the instability of PEMB during storage condition is a crucial limitation since the PE particles have a tendency to separate from the binder because the viscosity and the density of the binder different than these of PE polymer. In addition, there is no chemical compatibility between binder and PE. This lack of compatibility is occurred because PE polymers are nonpolar unlike asphalt binder, resulting in reducing the modification efficiency (Kakar et al., 2021; Revelli et al., 2024). In view of this, it is important for the polymer-modified binder to have good storage stability. At storage condition, the PEMB is considered stable when there are minimal differences between the properties of the top and base samples. Otherwise, a phase separation between asphalt binder and PE polymers has occurred in the PEMB. Two forms of phase separation were seen in the PEMBs without suitable storage stability. In the first form, the PE particles tend to float at the top of the tube surface due to their low density, which is lower than that of asphalt binder. In the second form, the PE tend to settle at the base of the tube due to the opposite reason. To confirm this, (Costa et al., 2013) revealed that HDPE and LDPE modified asphalt binders had poor storage stability comparing to other plastic polymers such as acrylonitrile–butadiene-styrene and crumb rubber which are more stable at storage condition. In addition, Ameri and Nasr (Ameri and Nasr, 2016) assessed the mixing process of devulcanized PE terephthalate and asphalt binder in terms of storage stability at high temperatures. The results indicated that the difference between the softening point of the top and bottom parts for all modified binders was higher than 2.2 °C which represent the minimum limit. This suggests that PEMBs cannot be considered as storage-stable blends, posing a challenge during storage, transportation, and application of such polymer-modified binders.

Poor storage conditions of PEMB, like storage at high temperature and for a lengthy period, has been confirmed to be the main factor that extremely influencing the deterioration of PE-modified asphalt roads. In particular, many investigations have been conducted to overcome the problem of phase separation in PEMB and make it more stable during storage. The storage stability deficiency for PEMB is enhanced with the addition of cross-linking agent and polyoctenamer polymer (Padhan and Sreeram, 2018) and besides this, addition of nano-silica and silicone coupling agent can avoid the separation between PE and asphalt binder which eventually enhanced the storage stability performance of PEMB (Arshadi and Taherkhani, 2024; Ma et al., 2023). Moreover, combine PE, for both types HDPE and LDPE, with other polymers such as polybilt and styrene butadiene styrene in modifying asphalt binder can achieve good storage stability (Wahhab et al., 2017). Furthermore, the combined of 0.15% sulphur and 1% of maleic anhydride-grafted PE are used to attain better stability at storage for LDPE polymer-modified binder (Nizamuddin et al., 2024).

Ultimately, preparing PEMB through applying wet mixing faces the risk of segregation when stored at high temperatures, as the polymer tends to agglomerate and rise to the top of the mixing vessel. This requires higher temperatures and longer time when reused during the production of asphalt pavement. The aforementioned repetition often leads to an increase the cost of modification due to greater fuel consumption and oxidation of the asphalt due to repeated reheating of the PEMB during preparation. Therefore, it has become necessary to find an alternative mixing method that ensures the easy processing of bitumen using polymer, the elimination of phase separation, and the reduction in modification costs. The previous positive effects are the motivation behind performing this research. Hence, the "semi-wet" process, new mixing method, is used purposely to minimize the negative impacts that occur in the wet process, which includes adding the PEMB directly to the hot aggregate without storing this bitumen. In this research, the effect of using semi-wet mixing (SWM) on the performance of PEMB-mixtures is evaluated in terms of mechanical properties, volumetric characteristics, indirect tensile strength (ITS), and moisture damage resistance.

MATERIALS

In this study, three major components were used in the composition of the asphalt mixture. The first component is bitumen, which is a binding material in the mixture and contributes to providing the necessary flexibility to resist cracks and erosion resulting from traffic loads and weather conditions. The bitumen was obtained from the oil refinery located in Dora city-Baghdad. The physical properties of bitumen such as penetration, specific gravity, ductility, softening point, and flash point were examined. The second component is aggregate, which consists of gravel, sand, and filler. The aggregate was carefully selected according to the requirements of the local specification for the Wearing Course type IIIA (State Corporation for Roads and Bridges, 2003), as it contributes to providing the necessary strength and durability for the asphalt mixture. The design gradation of aggregate is depicted in Figure 1. The necessary tests were carried out, such as the gradation test, bulk-specific gravity, apparent specific gravity, clay lumps, water absorption, and abrasion wear percentage. Meanwhile, the filler material is

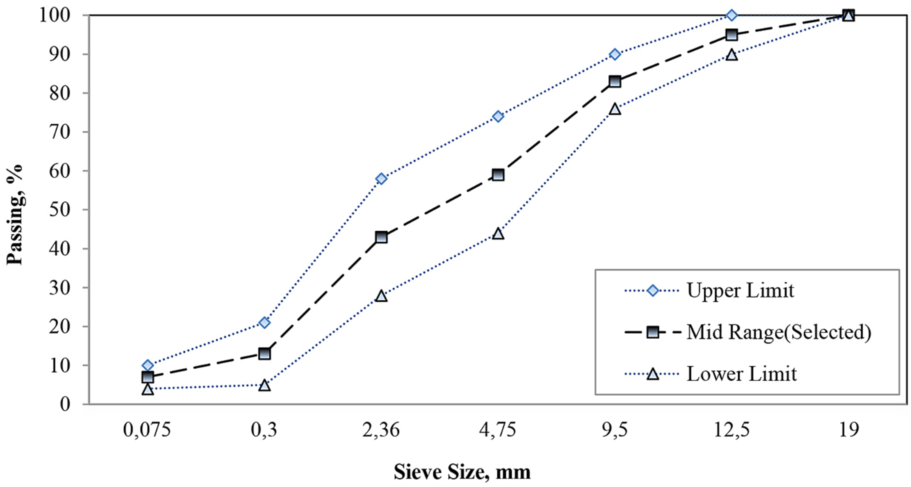

Figure 1. Design gradation curve for wearing course type IIIA

limestone that used in the research and has bulk specific gravity 2.71 and 87% of this filler is passed sieve No. 200 (opening size equal to 0.075 mm). Finally, the test results for both bitumen and aggregate are compared with the specification requirements (State Corporation for Roads and Bridges, 2003). The last component is PE polymer as shown in Figure 2. In this work, two kinds of PE were utilized as a bitumen modifying material; HDPE has density 0.954 gm/cm^3 and LDPE has density 0.924 gm/cm^3.

EXPERIMENTAL METHODS

Asphalt mixture preparation

The Marshall method is used in asphalt mixture design according to the ASTM D6927-15 specification to obtain the optimum bitumen content (OBC) in hot mix asphalt (HMA). Five bitumen percentages (4.0%, 4.5%, 5.0%, 5.5%, and 6.0%) were used, and three specimens were prepared for each percentage, for a total of fifteen specimens. Based on the bitumen percentage that achieves the highest stability, the percentage that gives the highest density, and the bitumen percentage that achieves voids of 4%, the required OBC was determined to be 4.9%. This percentage was employed in preparing all PE-modified mixtures to maintain consistency throughout the research period. To prepare the PE-modified mixtures, the components were mixed at temperatures between 160 and 170 °C and compacted at temperatures 10 °C lower than the corresponding mixing temperature (Attaelmanan et al., 2011). More attention must be given during mixing PE and binder through SWM because PE particles may be burn at too high temperatures and agglomerate at too low temperature. These detrimental impacts can lead to localized variations and uneven dispersion in modified mixtures.

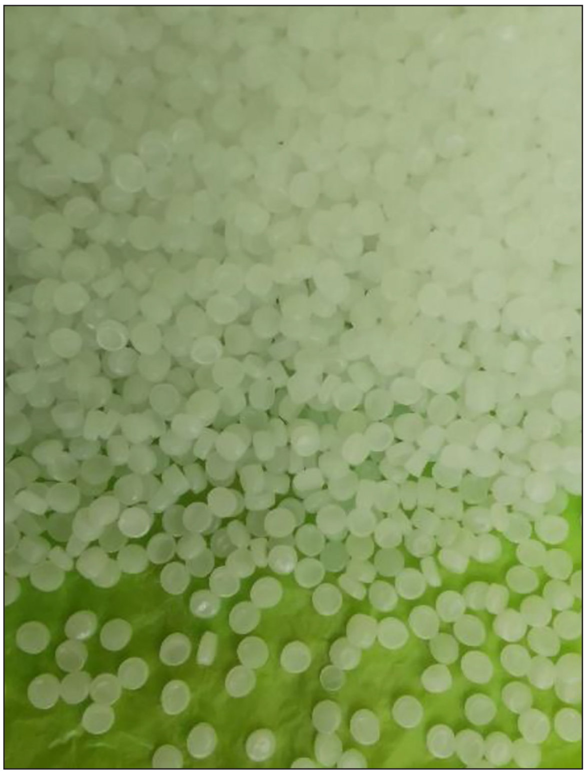

Figure 2. The morphology of the PE polymer

Using a Marshall press, the specimens were compacted of 75 blows per face of the mold. Then, the specimens were left in the mold to cure in air for 24 hours. Then, the mold was removed and the samples were treated in hot water bath at 60 °C before being examined using a Marshall apparatus. This device shall be designed to load at a uniform vertical movement 50 ± 5 mm/min. Marshall specimens testing procedure is illustrated in Figure 3.

Preparation of PE modified binder

Solid pellets PE polymer was mixed at three ratios 2, 4, and 6% by weight of OBC. More than 6% PE polymer in the bitumen could cause inhomogeneity and effect badly on the workability of the asphalt mixture. The bitumen to be modified is placed in a container covered with aluminium foil to prevent direct contact between the heat source and the bitumen container, where it is heated to a temperature of 163 °C, and then the PE is gradually added to the asphalt binder. SWM process was performed at a temperature of 165 °C, and for 30 minutes, it was used a high-speed stirrer rotating at 500 revolution per minute, resulting in a uniformly mixed bitumen. Figure 4 shows the PEMB.

Moisture damage test

Moisture sensitivity testing was performed according to ASTM D 4867M-96 on seven HMA sets categorized as follow: one set for standard mixture, three sets for HDPE-modified mixture and three sets for LDPE-modified mixture. Three Marshall samples were prepared for dry condition and three for wet condition for each set. These sets were compacted with air voids ranging from 6–8%. Tensile strength ratio (TSR) was computed

Figure 3. Marshall testing: (a) compacted sample, (b) Marshall device, (c) theoretical maximum specific gravity tester, (d) specimen fracture

based on the outcomes of the ITS test at 25 °C. The procedures of TSR testing are illustrated in Figure 5.

Figure 4. PEMB

RESULTS AND DISCUSSION

Marshall properties

Marshall Properties includes stability is an essential characteristic of asphalt concrete mixtures, since it indicates the mixture's capacity to withstand rutting. Increased stability of the asphalt mixture results in a higher level of rigidity, which in turn enhances its ability to withstand the pressure exerted by traffic loading (Mohammed et al., 2024). Obviously, the findings showed that the inclusion of 2%, 4%, and 6% of PE led to a respective increase in Marshall Stability of the asphalt mixture by 65.65%, 95.97% and 113.36% for HDPE, and by 48.26%, 67.82%, and 86.19%

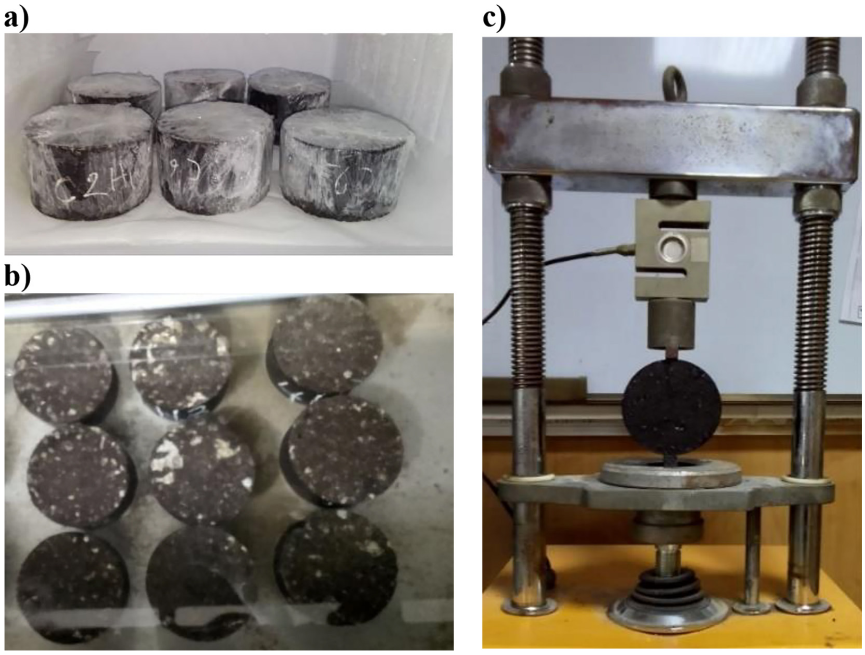

Figure 5. Moisture damage test: (a) freezing cycle, (b) thawing cycle, (c) specimen testing

for LDPE, respectively, compared to the standard mixture as shown in Figure 6. Another important parameter is flow. The high flow shows that the asphalt mixture exhibits minimal resistance to traffic loading under conventional conditions. A low value indicated that asphalt mixture is more rigid. As illustrated in Figure 7, the results of PE modified mix showed that the Marshall flow was reduced by 32.46%, 34.54%, and 34.8% with the addition of 2%, 4%, and 6% HDPE respectively. Meanwhile, the Marshall flow was reduced by 32.72%, 33.76%, and 39.74% with the addition of 2%, 4%, and 6% LDPE respectively, compared with the standard mixture.

The results reveal an increase in the stability value with raising the PE content in the modified mix. This is due to the increase in the PEMB viscosity with increasing PE content, which leads to enhanced bonding and interpenetration strength between binder and aggregate in the modified mix.

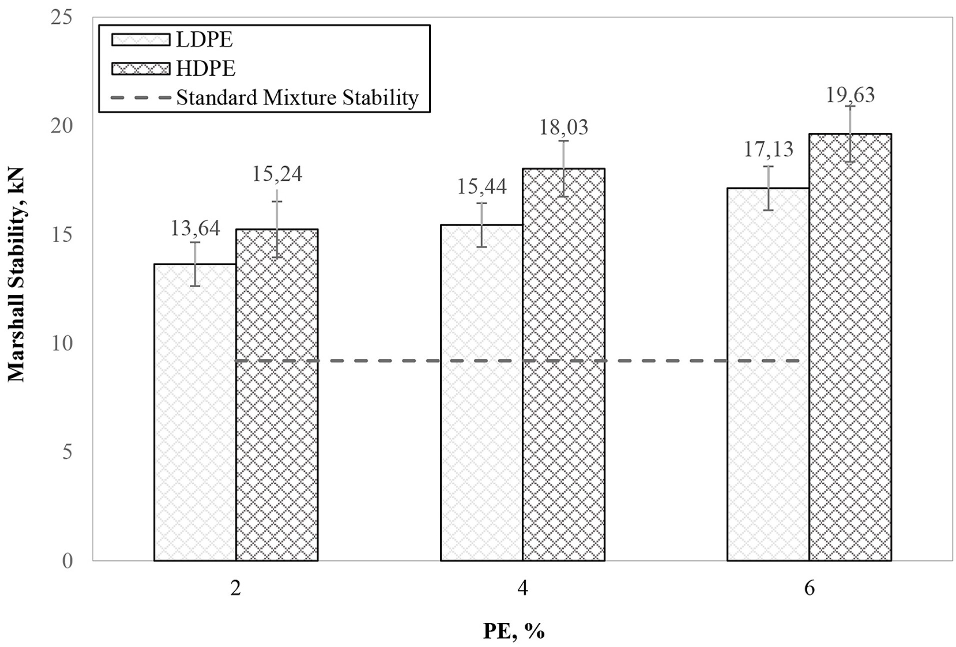

Figure 6. Marshall stability for standard and PE-modified mixtures

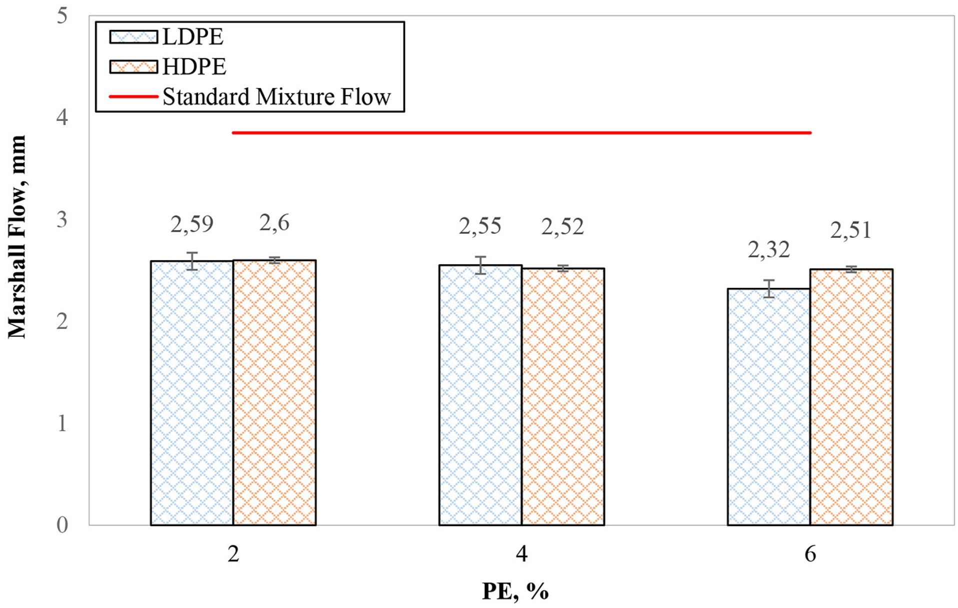

Figure 7. Marshall flow for standard and PE-modified mixtures

This in turn leads to a decrease in the flow value with increasing PE addition ration in the modified mixture compared to the standard mixture. Accordingly, the stability to flow ratio, known as the hardness index, increases with increasing PE polymer as shown in Table 1. This means that adding PE to asphalt mixes makes them stiffer and less affected by temperature.

The mechanical properties of PE-modified mixtures are enhanced due to the fact that SWM provide better compatibility and facilitate the homogenous dispersion of PE polymers (HDPE and LDPE) in bitumen. Eventually, adding PE to asphalt mixes is an enhancing factor to reduce the effect of permanent deformations that usually occur due to the change in binder viscoelastic properties at high temperatures. Therefore, using PE-modified binder reduces the possibility of paving problems that affect the overall performance of roads (Abduljabbar et al., 2022). As shown in Figure 8a, the density of all modified mixture was measured and compared with the standard mixture and a reduction in their density was noticed. This is due to the increase in the viscosity and lower density of the PEMB. Consequently, the percentage of air void in total mix and voids in mineral aggregate are increased as illustrated in Figure 8b and c, respectively. For that reason, the percentage of voids filled with bitumen was reduced and this can be seen in Fig. 8d. Nevertheless, for addition of 2% HDPE, the bulk density was increased and a reduction in voids in mineral aggregate was noted. This can be attributed to the low percentage of PE polymer. The outcomes are consistent with previous investigation (Lubis et al., 2020).

Furthermore, for standard and modified mixtures, the mechanical and volumetric properties met the local specification limit for the bitumen concrete mixtures (State Corporation for Roads and Bridges, 2003). The previously mentioned limits are listed in Table 2.

Table 1. Hardness index for standard and PE-modified mixtures

Mixture type	PE addition, %	Hardness index value, kN/mm
Standard mixture	0	2.389
LDPE modified mixture	2	5.26
	4	6.05
	6	7.38
HDPE modified mixture	2	5.86
	4	7.15
	6	7.82

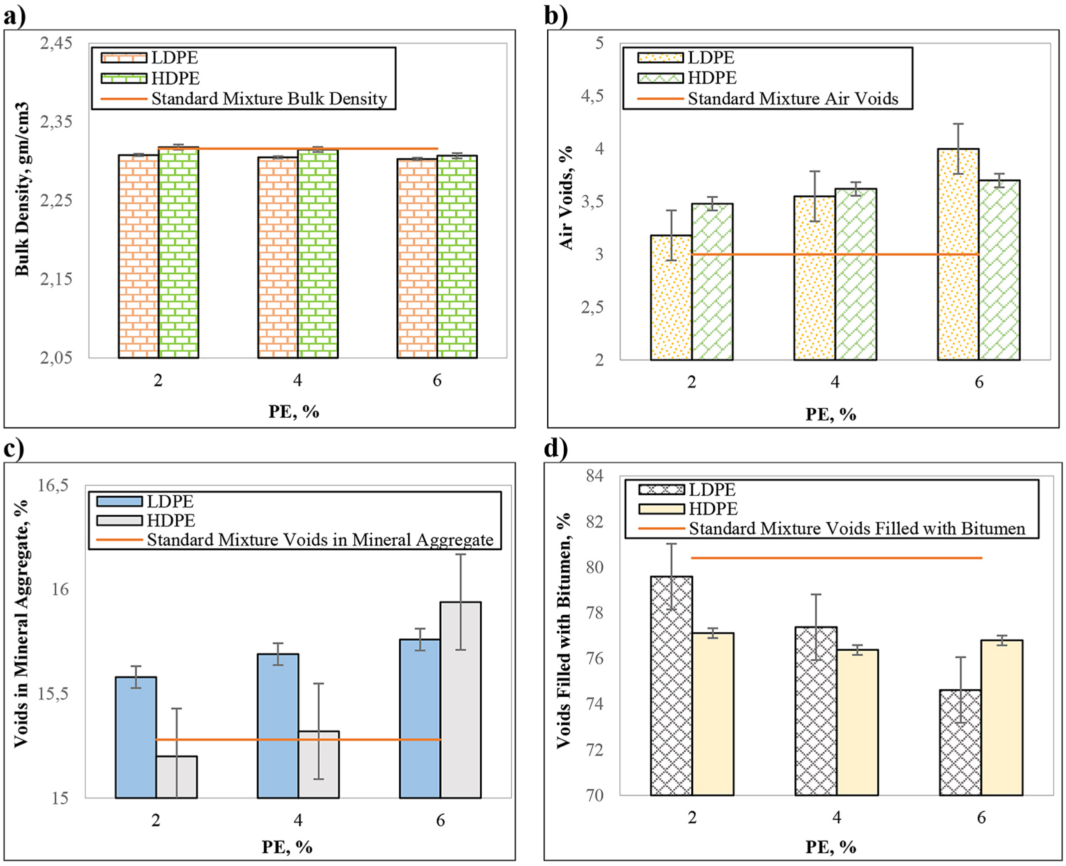

Figure 8. Bulk density and volumetric properties result for standard and PE-modified mixtures: (a) bulk density, (b) air voids, (c) voids in mineral aggregate, (d) voids filled with bitumen

Table 2. The mechanical and volumetric properties limit according to the specification for roads and bridges-section R9

Property	Unit	Specification limit (State Corporation for Roads and Bridges, 2003)
Marshall stability	kN	8 minimum
Marshall flow	mm	2–4
Hardness index	kN/mm	Not limited
Bulk density	gm/cm^3	Not limited
Air voids	%	3–5
Voids in mineral aggregate	%	14 minimum
Voids filled with bitumen	%	Not limited

Moisture damage assessment

Moisture has a substantial effect on the behaviour of asphalt mixes and can lead to deterioration of the mix over time. The negative effects of moisture include poor adhesion of the asphalt binder to the aggregate, resulting in reduced overall strength and durability of the asphalt mix (Ahmed et al., 2024). Rainwater is one of the primary reasons of moisture in HMA. Moisture trapped within the aggregate during the mixing process or during the paving stage can lead to subsequent problems, affecting the performance of asphalt mixes. These problems appear in the form of asphalt stripping, where moisture causes the asphalt to separate from the surface of the aggregate, reducing the cohesion between them. This phenomenon is known as "stripping". In addition, water accumulation within the asphalt mix is one of the causes of water erosion, which can lead to structural weakness and the formation of cracks and potholes. One of the suggested ways to address these problems is to use polymers to modify asphalt binder. Fig. 9 displays the ITS

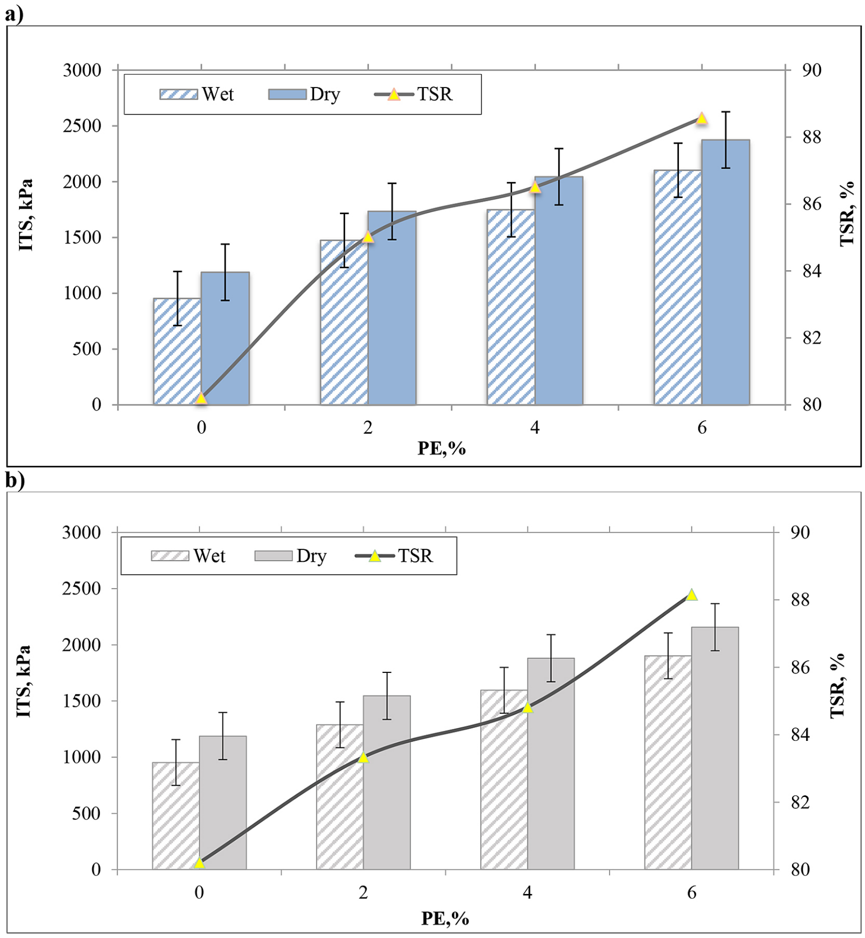

Figure 9. ITS and TSR for standard and PE-modified mixtures: (a) HDPE, (b) LDPE

and TSR results for the standard (0% PE) and PE-modified mixtures. The improvement can be seen with the addition of PE as an asphalt mix improver. For HDPE, the dry ITS results, as presented in Fig. 9a, increased by 45.9%, 72.09%, and 99.86% with additions of 2%, 4%, and 6%, respectively, compared to the standard mixture. Meanwhile, the wet ITS increased by 54.67%, 83.45%, and 120.69% with additions of 2%, 4%, and 6%, respectively. Also, as illustrated in Fig. 9b, the dry ITS increased by 30.1%, 58.32%, and 81.57% with additions of 2%, 4%, and 6% LDPE, respectively, compared to the standard mixture. Additionally, the wet ITS increased by 35.18%, 67.42%, and 99.58% with additions of 2%, 4%, and 6% of LDPE. To explain these outcomes, PE polymers can help in improving the adhesion and cohesion actions between the aggregates and the asphalt binder. Thus, the bond strength between PEMB and aggregate is increased, resulting in higher ITS and more resistance against cracking under tension

Obviously in Figure 9a and b, the outcomes of TSR when the addition of 2%, 4%, and 6% of HDPE resulted in an increase in TSR by 6%, 7.85%, and 10.42% respectively, and moreover, the addition of 2%, 4%, and 6% of LDPE resulted in an increase in TSR by 3.9%, 5.74%, and 9.91%, respectively. PE polymers can prevent the potential for moisture induced damage and stripping because the modification of binder with HDPE and LDPE enhance the shear binding between substrates and the bitumen. This will help in reducing the need for costly repairs and extending the lifespan of flexible roads. The TSR results are consistent with prior research (Abduljabbar et al., 2022).

CONCLUSIONS

The performance of PEMB-mixtures was assessed according to several indicators such as mechanical properties, volumetric properties, stiffness, tensile strength, and water damage resistance. HDPE and LDPE were used to modify the bitumen to improve the bonding and cohesion properties between the binder and the aggregate, resulting in a high-quality asphalt mixture that complies with specifications.

When PE was added through applying SWM, an improvement in Marshall properties and moisture resistance was observed with increasing PE content in the mixture. PE modification improves the ability of binder to sustain tensile stresses, besides, the initiation and propagation of cracks are prevented under these stresses. This enhancement can be attributed to the fact that PE modifies binder through forming a network of PE chains within the binder structure and that improves the PEMB elasticity and cohesion. Also, a rise in the hardness index of the PEMB mixture was noticed, indicating that PEMB mixture became highly resistant to temperature changes, in addition to its ability to withstand high and repeated traffic loads. As a results, these polymers will boost the overall performance of HMA, enhancing their quality and ability to withstand different environmental conditions.

From the previous results, it became clear that the mixtures modified with HDPE gave the best results compared to the mixtures modified with LDPE and the standard mixture that did not contain PE. In conclusion, PE polymer can be used as a modified bitumen using SWM that achieves positive results with speed of implementation and reduces the cost resulting from using the wet mixing. Thus, SWM is promising process and can be considered for use in preparing PEMB. It is recommended to modify the asphalt binder by performing SWM with others plastic polymers like, crumb rubber, polypropylene, and polyvinyl chloride.

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