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REVIEW ARTICLE

Sustainable Use of Polymer in Asphalt Mixture: A Review

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ABSTRACT – Polymer modified asphalt (PMA) mixtures are commonly employed in flexible pavement and overlays with large traffic volumes. In contrast, a number of laboratory and field tests have been conducted to evaluate the performance of PMA with that of conventional hot mix asphalt (HMA) mixtures. This paper presents an overview of the various polymers utilised in asphalt mixtures and their effects on the rheological, morphological, physical, and mechanical properties of polymer-modified asphalt binders. According to a review of the relevant literature, a good modifier affects the failure characteristics of the binder, allowing it to withstand greater stresses and strains before failing. In asphalt pavement, adding polymers to asphalt binders to improve their qualities has a long history. Due to polymers' limited solubility, polymer-modified asphalt binders may have several disadvantages. In addition, a brief description of the general approach to mixing polymers is provided. A summary of the benefits and drawbacks of the most common polymers used to alter asphalt binders Styrene-butadiene-styrene (SBS), Styrene-butadiene–rubber (SBR), Polyethylene Terephthalate (PET), Polypropylene (PP), Polyvinyl Chloride(PVC) etc. are illusrated. The paper continues with a discussion of the lasting effects of PMAs on asphalt pavement. To evaluate the economic and environmental impacts, a life cycle assessment (LCA) is required.

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INTRODUCTION

In recent years, the highway construction sector has expanded rapidly worldwide as traffic loads have increased [1]. The sustainability of asphalt pavement is a significant concern since it may lead to a crucial failure in roads and highways. The European Asphalt Pavement Association (EAPA) reports that Europe and the United States produced 265.4 million tonnes and 319 million tonnes of asphalt mixtures in 2014 [2]. The preceding has sparked interest in enhancing the performance of asphalt mixtures, their mechanical qualities, and road functionality. The asphalt binder is responsible for the flexibility of asphalt pavement, binding the aggregate together, and providing waterproofing qualities. Polymer addition to composite materials such as asphalt mixes has been studied [3,4]. Due to their excellent beneficial properties of the polymer, such as large surface area, remarkable dispersion ability, strong absorption, excellent stability, and high chemical purity, nanomaterial has recently attracted a lot of attention from pavement researchers for the preparation of durable asphaltic mixtures with high performance [1,5]. Polymer scientists and civil engineers have made numerous attempts to improve the performance of asphalt pavements. Researchers have recently studied to enhance binders with different additives such as natural and synthetic fibers, rubber, and polymers. Asphalt concrete pavements are still famous because they give excellent stability, longevity, and moisture damage resistance. The number of vehicles on the road is steadily increasing. This work aims to summarise the findings on the properties of asphalt mixes comprising various PMA additives such as fibers and polymers. This analysis will focus mostly on asphalt for road construction, as approximately 85 percent of asphalt is used in the pavement [6].

Elastomers and plastomers are two fundamental polymers often used to modify bituminous binders [7]. The term "polymer" simply refers to huge molecules produced by the chemical reaction of several tiny molecules (monomers) to form long chains. The physical qualities of a particular polymer are dictated by the sequence and chemical structure of the monomers it is composed of, as well as its molecular weight and molecular weight distribution.

Using polymer-modified binders is unquestionably more expensive than using unmodified binders. As a result, the performance improvements provided by PMA mixtures must be carefully assessed to compare the life-cycle costs of pavements and overlays using these mixtures to those using standard HMA mixtures [8]. Malaysia is closely monitoring global trends in plastic trash output and the use of single-use plastics and has been the world's largest importer of plastic waste since 2017 [9]. The use of waste materials such as polymers in pavement construction has garnered considerable interest in using waste polymer materials without compromising the pavement's quality.

History of Polymer used in Asphalt

Since 1843, synthetic and natural polymers have been utilized as a modifier in asphalt. The experiment began in Europe in the 1930s, and rubber latex was first used in North America in the 1950s. In the late 1970s, Europe adopted modified asphalts ahead of the United States, which was confined to using PMA due to its expensive costs [10,11]. The United States began to adopt newly created polymers and European technologies in the mid-1980s. Polymer-modified binders are currently incorporated in Australia's National Asphalt Specification [11] guidelines and specifications. In a

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1997 study of state transportation departments, 47 states claimed that modified asphalts would be used shortly, while 35 states said larger sections would be required. Several studies have been conducted worldwide to re-examine and analyze the benefits of altering polymers on pavement performance and work on defining requirements. Tests for binders are still ongoing [12].

PROPERTIES

Rheological Properties of PMAs

Fatigue cracking is caused primarily by applying stress-relaxation cycles caused by vehicle passing. Thermal cracking is caused by extreme changes in ambient temperature from high to low, resulting in almost evenly spaced cracks oriented orthogonally to the road direction. In contrast to rutting, thermal cracking is more likely in a cold climate because it causes the asphalt to become glassy, lowering the stress dissipation capability [13,14]. Furthermore, a good modifier alters the failure properties of the binder, allowing it to withstand higher stresses and strains before failing. The problems mentioned above are caused by the asphalt binders' complex rheological qualities. [15,16,17]. Asphalt binders have been shown to considerably impact the rutting and damage behavior of asphalt mixtures. Engineers and material scientists have attempted to modify the properties of binders using various modifiers such as fillers, fibers, extenders, plastics, anti-strip agents, oxidants, antioxidants, recovered rubber, polymers, and non-materials to overcome or minimize these difficulties. As demonstrated in Fig. 1 an ideal modifier would improve the rheological characteristics to resist the aforementioned distresses.

Furthermore, a good modifier alters the failure properties of the binder, allowing it to withstand higher stresses and strains before failing. Selecting an appropriate modifier to obtain the desired attributes can be helpful from an engineering and economic standpoint. Various polyolefins, natural rubber, thermoplastic elastomers, crumb rubber, and other modifiers have been researched, but only a few have proven adequate performance and cost. Asphalt binders are typically modified to improve one or more of the primary qualities of asphalt, such as stiffness, elasticity, brittleness, storage stability and durability, and resistance to cumulative damage.

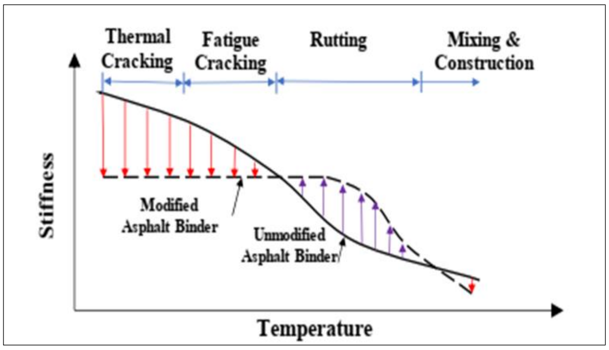


Figure 1. Expected change from modification in the rheological properties of asphalt binders [13].

Interaction of Polymer with Asphalt Phase

Among the four major classes of thermoplastic elastomers, it has been demonstrated that Styrenic block copolymers exhibit the most significant potential when combined with bitumen [18]. Styrenic block copolymers, commonly known as thermoplastic elastomers, can be synthesized sequentially by polymerizing Styrene butadiene styrene (SBS) or Styrene isoprene styrene (SIS) [19,20,21]. Alternatively, a diblock precursor can be synthesized sequentially by polymerizing styrene and a mid-block monomer, followed by a coupling agent reaction.

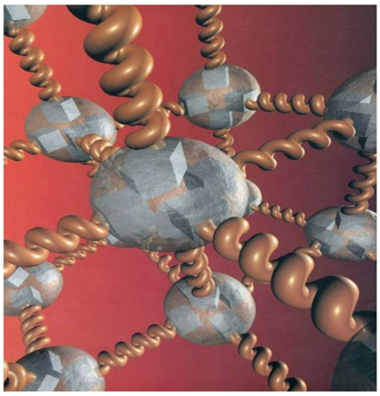


Figure 2. Schematic diagram of polymer molecules reacted with asphalt [18].

The strength and elasticity of thermoplastic elastomers are derived from the physical crosslinking of the molecules into a three-dimensional network. This is accomplished by aggregating the polystyrene end-blocks into distinct domains, as schematically depicted in Fig. 2, which serve as the physical crosslinks for a three-dimensional polybutadiene or polyisoprene rubbery matrix. The polystyrene end blocks provide the polymer's strength, while the polystyrene mid-block provides extraordinary elasticity [18]. Because the polystyrene (PS) end blocks aggregate into rigid domains at normal temperature, the polybutadiene (PB) or polyisoprene (PI) blocks operate as elastic amorphous unions between them, and SBS materials behave similarly to a crosslinked network [6]. Chemical modification of polymers is an efficient way to increase asphalt mix compatibility by improving their interaction with the asphalt phase [22,23]. Various techniques for improving the compatibility of asphalt blends through chemical modification have been proposed, including grafting reactive monomers onto polymers [24,25], functionalization of polymer with epoxy groups, acrylic acid, carboxylic acid, end amino, Glycidyl Methacrylate (GMA), Maleic Anhydride (MAH), and other reactive species/polymers [13], and the use of reactive species. When polymer chains interact and create chemical crosslinks, the chemical stabilization of reactive polymers happens. A hypothetical interaction between malted bitumen and Low-Density Polyethylene is depicted in Fig. 3 (LDPE).

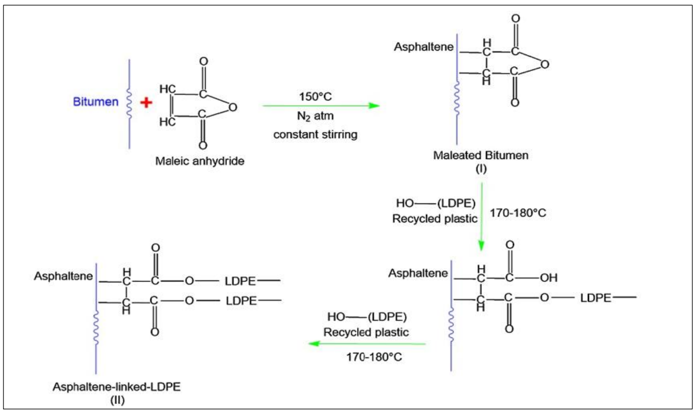


Figure 3. Hypothetical chemical interaction diagram of bitumen and LDPE [13].

Polymers Incorporated into Asphalt

Polymers used to modify bituminous binders can be classified into two broad categories: polymers and elastomers [7]. Plastomers typically modify bitumen by forming a challenging, rigid, three-dimensional network within the binder to provide resistance to deformation, whereas elastomers have a characteristically high elastic response and thus resist permanent deformation by stretching and recovering their initial shape. At the moment, the elastomer, styrene butadiene styrene (SBS), SEBS, and SBR styrene-butadiene–rubber (SBR) are the most often utilized polymers for bitumen modification. Plastomers are stiffer than thermoplastic elastomers, yet they have little reversible elasticity. It has a solid early strength under load; however, they have limited strain tolerance before brittle breakdown. Ethylene Vinyl Acetate (EVA), Ethylene Butyl Acrylate (EBA), Low-Density Polyethylene (LDPE), High-Density Polyethylene (HDPE), Ethylene-Propylene-Diene Monomer (EPDM), Ethylene Methacrylate (EMA), Polyethylene (PE), Polypropylene (PP), Polyvinyl Chloride (PVC), and Polystyrene are some examples of (PS) [13].

Thermoplastic Elastomers

Styrene-butadiene-styrene, SBS

Styrene-butadiene-styrene (SBS) is frequently utilized in asphalt pavements because it improves the upper, middle, and lower service temperature qualities [26,27,28]. Numerous researchers have demonstrated that asphalt mixes, including SBS-modified binders (SBS-MBs), exhibit increased resistance to permanent deformation, fatigue cracks, and damage caused by moisture penetration. Islam et al. found that when SBS content exceeds 3% by weight, the subsequent creation of an interconnected maltene-polymer-rich network significantly improves the properties of SBS-MBs. Even a modest modification to the polymer network in the steep intermediate zone (3 to 6% by weight) can dramatically alter the properties of SBS-MBs [29].

Styrene ethylene–co-butylene styrene, SEBS

SEBS is styrene (ethylene–co-butylene) styrene triblock copolymers formed by the simple hydrogenation of SBS. SEBS has a significantly lower polarity than SBS. The approximate solubility parameters for asphalt components and copolymer blocks. SEBS is miscible with maltenes but is nearly insoluble with asphaltenes. Becker et al. [30] discovered that SEBS-based PMAs constructed with a low penetration grade asphalt were more stable than SBS-based PMAs. SEBS has also been examined as an asphalt modification to develop impermeable membranes. Compatibility constraints are relaxed in this situation since the PMA is immediately cooled to room temperature following the mixing method, allowing for the freezing of the shear-induced metastable morphology [31].

Styrene-butadiene–rubber, SBR

SBR (styrene-butadiene–rubber) has been one of the most effective asphalt modifiers for paving asphalt. The layered silicate comprises layers of tetrahedral silicate sheets and octahedral hydroxide sheets. It is well recognized for its thermal stability and low cost [5]. Montmorillonite (MMT), vermiculite (VMT), rectorite (REC), and kaolinite clay are the primary components (KC). Polymer chains can intercalate into the interlayer of silicate in layered silicate modified polymers, causing the silicate to be disseminated throughout the polymer matrix at the nanoscale scale, resulting in considerable improvements in thermal and mechanical and barrier properties [32]. Zhang et al. [33] studied that By integrating MMT into styrene-butadiene–rubber and mixing it with asphalt, modified styrene-butadiene–rubber/montmorillonite (SBR/MMT) asphalt was made. Its characteristics and properties were determined through the use of traditional test methods. The X-ray diffraction (XRD) analysis results indicate that SBR/MMT composites can produce an intercalated structure, whereas SBR/MMT modified asphalts can generate an exfoliated structure. Fluorescent images demonstrate that modified asphalts containing SBR/MMT created an excellent fine network structure. SBR/MMT additions improved the physical qualities of asphalts by increasing their softening point and viscosity and decreasing their penetration at elevated temperatures.

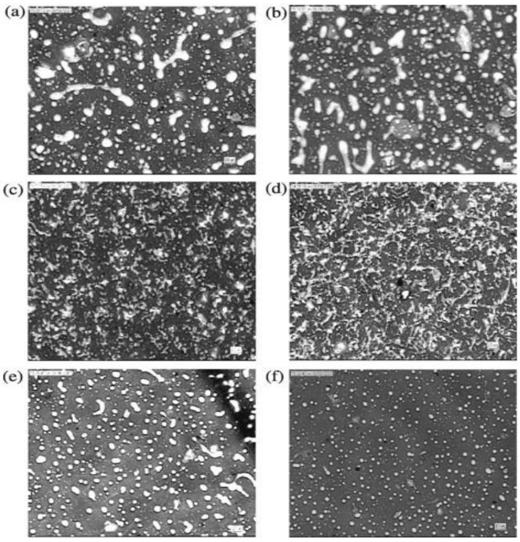


Figure 4: Fluorescent images for modified asphalts using SBR/MMT with different ratio [33].

Plastomers

Polyethylene terephthalate, PET

PET is a thermoplastic polymer; it is a kind of resin of the polyester family made by polymerizing ethylene glycol and terephthalic acid. It is widely used to make plastic bottles. The plastic with the most significant market production is polyethylene, accounting for 34% of the plastics market. PE is split into two types: LDPE, which has a density between 0.91 and 0.94, and HDPE, which has a thickness greater than 0.94 and is produced by polymerization at high and low pressures, respectively [34]. PE is a thermoplastic polymer that can be molded at a high temperature but cools to a rigid, solid state. However, thermosetting materials, such as crosslinked polyethylene, can be added. PE for wet asphalt modification [35,36] was the subject of the initial research. With a proportion of PE, the mixing temperature ranges between 150 and 180 °C. When the asphalt binder softens at high pavement service temperatures, the PE particles remain solid, improving the rutting resistance of the modified binder blends. On a microscale, specific plastic components may interact with asphalt to form a network that stops the molecular chain of asphalt from moving [37]. Al-Haydari et al. [38] examined a substitute 7.5 % asphalt binder using PET, increased mixture stability by 10%, and reduced flow by approximately 26% compared to a typical mix. Utilizing waste PET up to 10% by weight of asphalt results in an increase in bulk density and VFA and a cost reduction. With the growing concern for environmental protection, the highway industry recycles PET waste by using it as an additive in asphalt concrete or as an acceptable aggregate replacement. PET can be added using both a dry and a wet method.

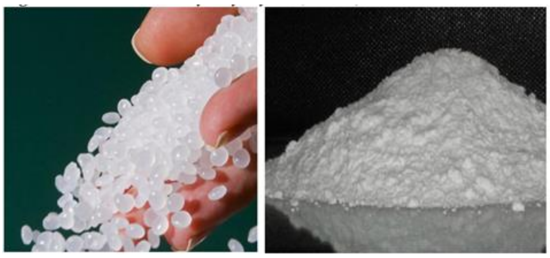


Figure 4: Polyethylene (LLDPE) as granular and powder state [39].

Polypropylene, PP

Polypropylene (PP) is a thermoplastic linear hydrocarbon with an intermediate crystalline level between LDPE and HDPE. PP-modified asphalt mixtures have shown reduced crack intensities while not being effective in reducing or delaying deflection cracking. Implementing asphalt mixtures with PP through either the wet or dry methods improves. Adding PP to the asphalt system could reduce the brittleness of the asphalt mixture at low temperatures and enhance its flexibility at high temperatures. At a volume fraction of 0.5 percent, PP fibers improved Marshall Stability, indirect tensile strength, and moisture susceptibility, according to [40]. Abtahi et al. [41] discovered that PP modified asphalt concrete helps the asphalt concrete mixture operate better. According to the findings, Marshall Stability and percent air void rise when flow property diminishes.

Regarding Marshall, indirect tensile strength, and compressive strength, AlHadidy and Yi-Qiu [42] concluded that PP-modified asphalt mixtures outperformed typical mixtures. On the other hand, the temperature susceptibility was reduced after adding PP to the asphalt mixture [43]. Most polypropylene used is highly crystalline and regular (i.e. isotactic), opposite to amorphous thermotics, such as polystyrene, PVC, polyamide, etc., where radicals are placed randomly (i.e., atactic).

Polyvinyl chloride, PVC

Polyvinyl Chloride, PVC has a high softening temperature of about (100-260) °C, raising the asphalt binder's softening point [44]. Also, the softening point is related to the fast connectivity of particles, especially when the temperature is raised, which leads to increased cohesion between the chains linked to each other and, therefore, higher resistance to permanent deformation. Furthermore, adding PVC to the binder enhances its viscosity. PVC causes a significant improvement in bitumen characteristics by lowering the penetration grade and increasing the softening point. Compared to unmodified samples, it is also less sensitive to temperature variations and more resistant to rutting at high temperatures, which is desirable [45]. PVC powder causes a significant improvement in bitumen characteristics by increasing the softening point and lowering the penetration grade.

In comparison to unmodified samples, it is also less sensitive to temperature variations and more resistant to rutting at high temperatures, which is desirable. Arabani et al. show the addition of 5% PVC powder to HMA mixtures increases their static performance [46]. It may be determined that mixes containing waste PVC powder are more resistant to permanent deformation when the MQ parameter increases.

Summary of the Influence Factors on PMAs

Table 1. There are some shortlisted polymers are described [6,13,29, 44,47,48].

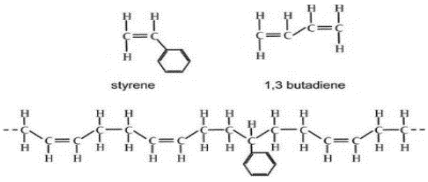
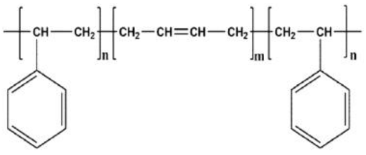
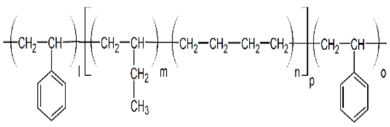
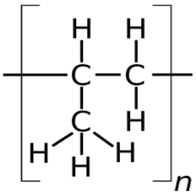
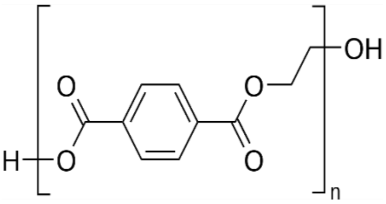
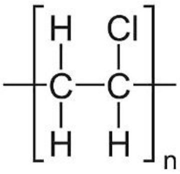
Polymer	Chemical structure	Advantages	Disadvantages
		Plastomers	
SBR		Excellent aging stability Resistance to thermal cracking has been improved Temperature susceptibility is reduced Increased viscous flow resistance Cohesion has improved Resistance to thermal cracking has been improved.	Low anti-aging resistance Problems with compatibility
SBS		Increased viscous flow resistance Improved thermal crack resistance Improved cohesiveness Good aging stability Reduced temperature susceptibility Increased thermal crack resistance	Cost is relatively expensive Some binders have compatibility issues. Compared to specific polymers, such as polyolefins, it has a lower resilience to heat, oxidation, and UV radiation
SEBS		Oxidation, Ultraviolet and high resistance to heat	Relatively high cost Storage stability problems Reduced elasticity compared to SBS's

Table 1. There are some shortlisted polymers are described [6,13,29, 44,47,48] (cont.)

Polymer	Chemical structure	Advantages	Disadvantages
		Elastomers	
PP		Enhance high-temperature performance Costs are relatively modest. Widening the plasticity range improved load resistance. Temperature susceptibility has improved.	Problems with phase separation in asphalt High crystallization potential Improvement in elastic recovery is negligible. Resistance to thermal fatigue cracking is low. Variations in fundamental binder characterization tests
PET		Enhance high-temperature performance Rutting resistance is high, Rutting resistance is high, Increased modulus at a relatively modest cost Fracture toughness has improved.	Phase separation and instability are challenging to combine in asphalt. Dangling modifier, Effective polymer content is required. Improvement in elastic recovery is negligible. At low temperatures, performance suffers
PVC		PVC disposal with minor cracking Improves performance of HMA	Principally serves as a filler character

EXPERIMENT WITH FACTORIAL AND FIELD TEST SECTIONS

According to the Standard specification for road works, Jabatan Kerja Raya Malaysia (JKR), a PG 76 or greater performance grade shall be achieved by adding sufficient polymer additives to traditional bitumen with grade 70-100 by MS 124. The polymer must be noncancerous [49]. They supplied the suggested polymer modified binder's (PMB) material safety data sheet. The polymer-modified binder (PMB) shall have the qualities specified in Table 2.

Table 2. Properties of polymer-modified binder [49].

Test	Requirement	Test specification
PMB before Rolling Thin Flim Oven Test(RTFOT)		
Viscosity, max.3 Pa.s,Test temperature °C	135	ASTM D 4402
Dynamic shear, $G/\sin \delta$ minimum 1.00 kPa, 10 rad/s, test temp °C	76	AASHTO T 315
Penetration, 100g, 5s , 25 °C, 0.1 mm	Report	ASTM D 5
Ring and ball softening point, minimum, °C	60	ASTM D 36
Flashpoint, minimum, °C	230	AASHTO T 48
Moisture sensitivity test, minimum, %	80	AASHTO T 283
Dynamic shear, $G/\sin \delta$ minimum 2.20 kPa, 10 rad/s, test temperature °C	76	AASHTO T 315

Mixing Method of PMAs

Polymers are divided into passive and active polymers [50]. Asphalt is treated with passive polymers before being physically blended. Chemical crosslinks are formed when active polymers react chemically with asphalt. These polymers' reactivity is due to functional groups reportedly capable of bonding with asphalt molecules. Passive polymers include materials like SBS and EVA, which are commonly utilized. Active polymers include ethylene terpolymers and Glycidyl Methacrylate (GMA) [13].

There are two methods for incorporating recovered plastics into asphalt mixtures: wet and dry. Fig. 4 illustrates the wet and dry processes used to incorporate polymers into asphalt plants [36]. The wet process begins with combining polymers and bitumen at elevated temperatures to create bituminous binders mixed with aggregate. As a result, the wet process necessitates the addition of machinery and equipment to shred the polymers into powders and then combine them with a hot asphalt binder [51]. The wet process of blending plastics with asphalt is a physical method in which plastic particles absorb the lightweight components of asphalt to generate a viscoelastic phase at elevated temperatures. This technique is suitable for plastics with relatively low melting points, mainly PE [12]. The dry method incorporates polymers or plastics as an aggregate replacement or mixture modifiers straight into the mixture. The aggregate replacement approach is frequently used with plastics with a high melting point, such as PET and PS.

In contrast, the mixture modifier approach is used with virtually all types of recycled plastics (e.g., PE, PP, PET), except polyvinyl chloride (PVC), due to concerns about hazardous chloride emissions [52,53]. When polymers with a melting point lower than the temperature at which the combination is produced are used to modify the mixture via the dry process, they melt when mixed with the hot aggregates, resulting in plastic-coated totals with possibly better physical and surface properties. As a result, the dry process appeared easy and energy-efficient by simply incorporating the shredded plastic granules into heated aggregates.

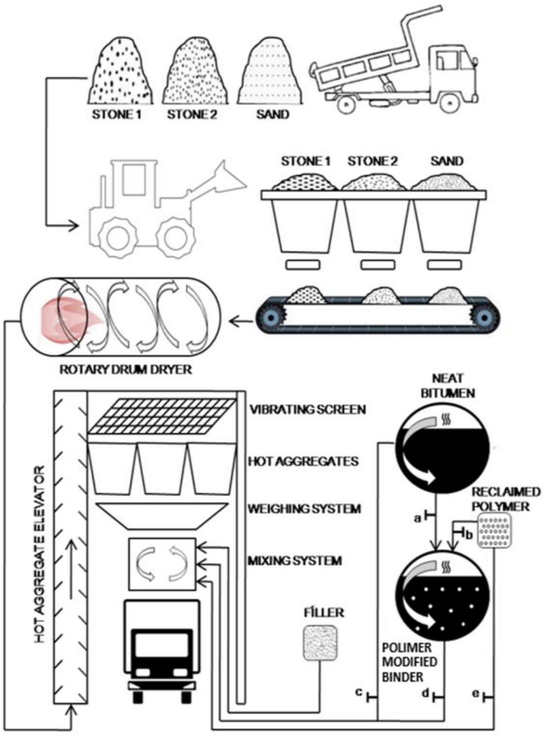


Figure 4. PMA mixing process in a discontinuous asphalt plant [54].

SUSTAINABLE IMPACT OF PMAS

Modified binders are frequently employed in high-stress, high-traffic, and extreme-climate environments [55]. Engineers can select suitable modifiers to obtain the desired qualities by understanding the effects of modifiers on the performance of asphalt binders. Furthermore, quality control limitations for modified asphalt binder might be established. To gain better performance, attributes of polymer additives, such as composition and molar mass, could be changed. To measure the apparent increase in life afforded by pavements with PMA mixes, it appears appropriate to combine the body of evidence in this area. It would also be beneficial to discover the site and design elements (such as traffic, climate, thickness, and so on) that can maximize the benefit of employing PMA.

Additives generally exhibit a phenomenon known as pseudo-plasticity, in which viscosity values are dependent on shear rate. As a result, binder test procedures may not be appropriate with changed binders [55] [45]. Polymer content ranges between 2 and 10% by weight; however, the most common proportions were about 5 or 6% a few years ago. Now the polymer content has been reduced to 2 or 3%. Sometimes waste materials (due to their low cost, they might be added in higher proportions) or mixes of two different polymers (as mentioned before) are used. The qualities of modified asphalt binders are determined by the neat features, the added modifier's attributes, mixing conditions, and the modifier's compatibility with asphalt [13]. The service life extension of PMA combinations over conventional, unmodified mixtures was highly variable, ranging from five to more than ten years [8]. This equates to a 20% to 100% improvement in performance, assuming a typical pavement life cycle produced with traditional HMA mixes is ten years before the first significant rehabilitation is done.

The usage of different plastics in asphalt pavement, life cycle cost analysis (LCCA), and life cycle assessment (LCA) are required to quantify the economic and environmental implications. Poulikakos et al.[56] did an LCA in road building utilizing PET and glass. They discovered that the cost of materials, greenhouse gas emissions, and energy savings were 8.5%-33.9 %, 26.63%, 86 %, and 13.9 %, 76.1%, respectively [56]. The production costs were calculated using the raw material prices. Poulikakos et al. and other researchers explained that the cost savings from preventing waste disposal were computed using the difference between treated waste and raw material prices. For example, avoiding the removal of crumb rubber in municipal garbage incineration [52] results in significant CO_2 emissions savings in one of the products that use CR [57].

CONCLUSIONS AND RECOMMENDATIONS

The quality of its road networks influences every society's economic progress. Asphalt pavements are made up of asphalt binders, aggregates, and additives and are one of the essential components of road networks. Asphalt binder's rheological and mechanical qualities significantly impact how well asphalt mixes operate over time. Polymers can improve the viscosity of asphalt binders at high temperatures, whereas, at low temperatures, they can increase their elasticity. A good modifier alters the failure qualities of the binder, allowing it to withstand higher stresses and strains before failing. To obtain the necessary attributes, selecting an appropriate polymer for modification can be effective from an engineering and economic standpoint.

Moreover, the modified polymer must be cost-effective; the polymer should improve the rheology and strength of the asphalt mixed with it. Any increased road costs imposed by its use are recovered through performance and reduced resurfacing costs. Many PMA pavements had very little fatigue cracking compared to their comparable control sections with unmodified mixtures, one of the more relevant discoveries from the recent field experiments. The results of laboratory fatigue tests and accelerated pavement tests are beginning to be confirmed in these test areas. Selecting an appropriate modifier to obtain the desired attributes can be helpful from an engineering and economic standpoint.

However, two recommended points should be considered concerning that assertion. The 5- to 10-year period is conservative or low because many PMA test sections have not yet shown any fatigue or load-related cracking. – Second, because some of the standard unmodified HMA mixtures employed as companion test sections represent Marshall and Hveem mixture design technologies, the 5 to 10-year estimate could be conservative. Furthermore, further study is required to develop novel recyclable and environmentally acceptable polymer modifiers. One of the critical objectives of future research should be the viability of using various waste/recycled and biobased materials and their impact on the rheological and physical properties of asphalt binders and mixtures.

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