Some Initial Results of the Research on Using Recycled Polyethylene to Modify Bitumen

Tran Viet Khanh, Nguyen Ba Hoang, and Vo Dai Tu

Abstract Asphalt concrete (AC) has long been a popular material used as a pavement texture, however, there are still limitations in the exploitation process. Bitumen has an important role, it has a great influence on the properties of AC, improving bitumen quality is a trend in improving AC quality. Besides some currently used such as rubber additives, sulfur additives, polymer additives, finding new types of additives with easy, cost effective supply, taking advantage of recyclable material is always a matter of necessity. This paper presents some initial results of the research on using recycled polyethylene (RPE) to improve bitumen. This study introduces the mechanism of interaction between polyethylene (PE) and bitumen and conducts experimental studies to evaluate the influence of PE/RPE on the properties of bitumen. PE/RPE with different contents is added to 60/70 bitumen to analyze the parameters of bitumen such as penetration, softening point, ductility, elastic recovery, adhesion to stone, viscosity, storage stability. The results of the study to initially assess the influence of PE/RPE additives on the specifications of the bitumen, it is a premise for other studies in using PE/RPE to create PE improved bitumen (PEMB), RPE modified bitumen (RPEMB), creates new AC with better quality.

Keywords Additives · Modified bitumen · Polymer modified bitumen · Polyethylene · Recycle polyethylene

1 Introduction

Improving the quality of AC is always a necessary issue in the world as well as in Vietnam. Besides improving the quality of design and construction, the use of modified bitumen is a very effective solution to improve the quality of AC. The use of polymer additives to improve bitumen (PMB-Polymer Modified Bitumen), to

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a) PE b) Products of PE c) Wastes from PE

Fig. 1 PE, products and wastes from PE

produce polymer AC (PMA-Polymer Modified Asphalt concrete) has been studied and applied in many parts of the world, with outstanding results compared with traditional AC. PMA reduces the ability to soften at elevated temperatures (up to 90 °C), less prone to cracking at low temperatures (down to −40 °C), so it is higher stiffness in high temperatures, lower stiffness in low temperatures, improve resistance to wheel rutting, improve resistance to cracking and fatigue, increase road surface roughness and service life many times longer than AC [1–4]. Some polymer additives have been studied and applied such as: SBS (styrene-butadiene-styrene), SBR (styrene-butadiene-rubber), EVA (ethylene-vinyl acetate), … The technology to produce PMB by mixing bitumen with thermoplastic polymer has been adopted and is widely used with production of millions of tons/years [5, 6].

PE is a thermoplastic polymer with high strength, PE products are plastic bags, home appliances, industrial plastic equipment, they are very popular (Fig. 1).

Using PE to improve bitumen has been studied since the 2000s and has recently given very positive results. Using PE will reduce the penetration, reduce the elongation of the bitumen, increase the softening temperature, increase the ignition temperature, and increase the viscosity of the bitumen [7–9]. PE will increase the shear modulus, reduce phase angle and increase the rotational viscosity of the bitumen, make the bitumen improve its elasticity and increase the wheel rutting resistance for AC [10]. Studies [8, 11] show that AC using PEMB will significantly improve the stability, slightly reduce the flow and bulk density of AC, the air voids of AC will not increase significantly, so PE will reduce deformation of AC, increase fatigue resistance and create better bond between asphalt and aggregate. AC using PEMB also reduces the dynamic deformation and reduces the wheel rutting of significantly [9].

Nowadays, the overuse of plastic products from PE is causing serious harm to the environment while PE is recyclable for use. So, the study of using PE/RPE to improve bitumen and AC quality has scientific and practical significance, helping to find a new additive to improve bitumen with available supply, make use of recycled materials, save costs and contribute to reducing environmental pollution.

2 Interaction Between PE and Bitumen

2.1 Interaction Between PE and Bitumen

Bitumen is a mixture with the complex chemical composition, however, it is possible to separate bitumen into two broad chemical groups called asphaltenes and maltenes. The maltenes can be further subdivided into saturates, aromatics and resins. The four groups are not well defined and there is some overlap between the groups.

Bitumen is regarded as a colloidal system consisting of asphaltene micelles dispersed or dissolved in the oily medium (maltenes). The micelles are considered to be asphaltenes together with an absorbed sheath of high molecular weight aromatic resins which act as a stability solvating layer. In the presence of sufficient quantities of resins and aromatics of adequate solvating power, the asphaltenes are fully peptised and the resulting micelles have good mobility within the bitumen. These are known as 'SOL' type bitumen (Fig. 2a). If the aromatic/resin fraction is not present in sufficient quantities to peptise the micelles, or has insufficient solvating power, the asphaltenes can associate together further. This can lead to an irregular open packed structure of linked micelles in which the internal voids are filled with a fluid with a mixed composition. These types of bitumen are known as 'GEL' types (Fig. 2b) [12]. In practice, most types of bitumen are of intermediate character.

PE is a polymeric organic compound, prepared from the polymerization of ethylene monomers (–CH_2–CH_2–) (Fig. 3). PE does not react with acid solutions and alkaline solutions, insoluble in water and alcohols, PE poorly dissolves in solvents such as toluene, trichlorethylene at temperatures higher than 70 °C. PE is stable with temperature and has good elasticity, mechanical toughness, high tear strength, good abrasion resistance and is durable against chemical agents.

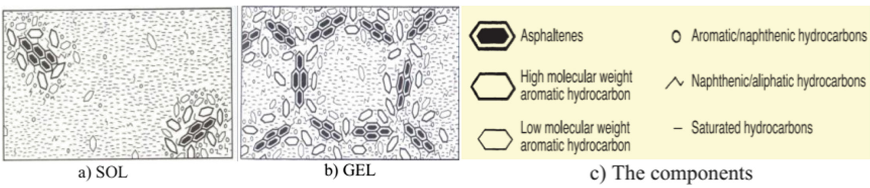

Fig. 2 Structure of bitumen

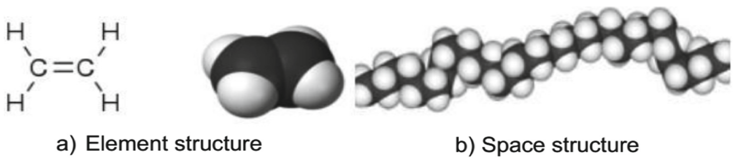

Fig. 3 Structure of PE

When adding a small amount of PE additive to bitumen, even when mixed at high temperature (above 160 °C), PE is insoluble in bitumen, there is no chemical reaction between PE and bitumen, the PE particles (crushed) exist as a dispersant in the bitumen-PE colloidal system (PEMB). The PEMB colloidal system is a multi-dispersion system, the dispersing medium is maltenes, the dispersant is the asphaltenes and the PE (crushed). The stability of the PEMB multi-dispersion system depends on whether the asphaltenes and PE dispersants are evenly dispersed in the PEMB system over time, depending on the PE content, resins and aromatics content in bitumen and the temperature of the PEMB system. If the content of resins and aromatics is not enough, at high temperature, the asphaltenes tend to bind together and settle more quickly downwards, the PE particles tend to float above and to bind together. This makes the PEMB colloidal system tend to be unstable, separated during storage. High resin and aromatics content is more helpful in balancing the PEMB system.

2.2 Blending PE/RPE with Bitumen

Equipment used to mix Bitumen and PE/RPE is shown in Fig. 4. The main parts are motor, rotating shaft, mixing wings, mixing barrel, temperature regulator, rotation speed regulator. We can mix 2 mixing barrels at the same time. The mixing process can be summarized through the steps:

Step 1: Heat bitumen in the drying cabinet to 150–160 °C, pour bitumen into mixing barrel, weigh and record the bitumen mass;

Step 2: Calculate the amount of PE/RPE additive to use (% by bitumen mass); spread PE/RPE evenly onto the bitumen surface in the mixing barrel;

Step 3: Place the mixing barrel (with bitumen and PE/RPE) on the mixer so that the mixing wings is about 1cm from the bottom of the mixing barrel;

Step 4: Mixing.

+) Mixing speed: 800–900 rpm for the first 30 min, then 500–600 rpm;

+) Mixing temperature: maintain sample temperature during mixing of 165–170 °C (for PE) and 175–180 °C (for RPE);

+) Mixing time: about 1.5 h for PE and 2.0 h for RPE;

Step 5: Turn off the mixer, pour the sample into the container, and put it in a drying cabinet to release air bubbles before testing technical properties.

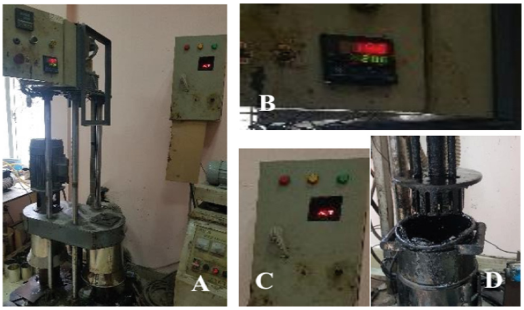

Fig. 4 Mixing equipment. **a** Mixing equipment. **b** Control the temperature. **c** Control the rotation speed. **d** Mixing wings and mixing barrel

Table 1 Testing properties

Properties	Test method
Penetration	TCVN 7495:2005
Softening point	TCVN 7497:2005
Ductility at 25 °C	TCVN 7496:2005
Elastic recovery	22TCN 319-04
Adhesion to stone	TCVN 7504:2005
Viscosity at 135 °C	22TCN 319-04
Storage stability	22TCN 319-04

3 Results of Empirical Research

3.1 Methodology

The test program is performed according to the following steps:

- Prepare material sources: PE/RPE, bitumen 60/70 with defined specifications.
- Design the modified bitumen mixtures, PE/RPE content added to bitumen 60/70 is 0, 2, 4, 6, 8% respectively of the mass of bitumen.
- Mix PE/RPE with bitumen 60/70 in a mixer to create modified bitumen samples, according to the components designed.
- Testing technical parameters of samples, listed in Table 1.
- Analyze, discuss about test results.

3.2 Materials

Bitumen: Bitumen 60/70 with Exxon brand from BMT Construction Investment Joint Stock Company, the properties are shown in Table 2.

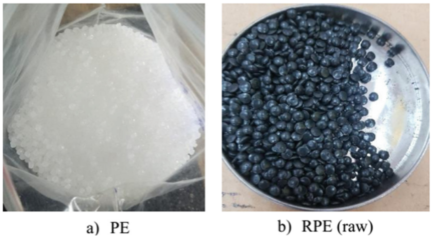

Fig. 5 Granular PE

Table 2 Properties of the bitumen 60/70

Properties	Unit	Result
Penetration	0.1 mm	65
Softening point	°C	48.6
Flash point	°C	368
Ductility at 25 °C	mm	131
Viscosity at 135 °C	cP	400
Specific gravity	g/cm^3	1.036
Solubility in trichloroethylene	%	>99.9

Table 3 Properties of PE

Properties	Unit	Result
Specific gravity	g/cm^3	0.924
Softening point	°C	93
Melting temperature	°C	110
Tensile strength at yield	N/mm2	11
Tensile modulus	N/mm2	260

The additive PE/RPE: PTT brand virgin PE and rough recycling PE from A Dong ADG JSC (Fig. 5), the parameters of PE are shown in Table 3.

3.3 Test Results and Evaluation

When the content of PE/RPE added to bitumen 60/70 increased from 0 to 2, 4, 6, 8%, the test results of the bitumen specifications changed as follows:

The penetration decreased from 65 to 32 (reduced to 49.2%) for PEMB, and decreased from 65 to 25 (reduced to 38.5%) for RPEMB, it's been reduced quite a lot (Fig. 6). This is because PE is tough, good tear strength, so it makes bitumen becomes denser. The penetration of RPEMB is much lower than that of PEMB because there are many impurities in the RPE, making modified bitumen harder. Reduced the penetration will make the bitumen harder, better anti-oxidation, improve the stability, resistance to abrasion, deformation and wheel rutting. However, if the penetration is too small, the bitumen will be too hard, will become more brittle, reduce elongation and intrinsic adhesion ability, reduce elasticity, make the bitumen more prone to cracking and fatigue damage. With this test result, the additive content used should not be more than 6%.

The softening point increases from 48.6 to 61.6 °C for PEMB, it increases by 26.7%, obvious but not too high. For PREMB it increases very high, above 100 °C with 8% of RPE (Fig. 7). This is because PE has a high softening point, good thermal stability. The softening point of RPEMB is higher than that of PEMB because the composition of RPE also contains more hard plastics, many impurities, make RPEMB harder. The increase in softening point will make the bitumen more stable with high temperature, improving the resistance to deformation, resistance to rutting, no plastic melt, material (in AC) not pushing during use. Bitumen is less sensitive but more stable under the influence of heat. However, if the softening point is too high, the mixing and compacting temperature will also increase, causing difficulties in the production and construction process. The additive content should not be more than 6%.

The elongation decreased from 131 to 119, 95, 84 cm (down to 90.8, 72.5, 64%), plummeted to 30 cm (down to 23%) with 8% of PE. For PREMB, the elongation is greatly reduced, down to 54.2, 31.3, 9.1, 4.6% (Fig. 8). This may be because PE has the property of hardening at low temperatures, making the bitumen tend to be stiffer. The elongation using RPE is reduced more than when using PE, because in the composition of RPE there are more hard plastics, more impurities make RPEMB

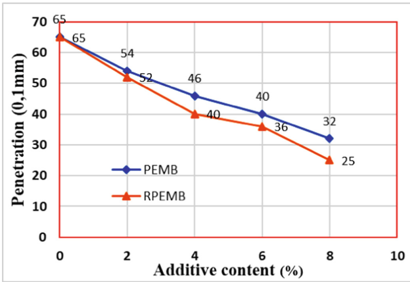

Fig. 6 Test result of penetration

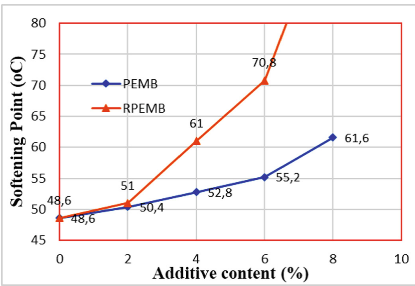

Fig. 7 Test result of softening point

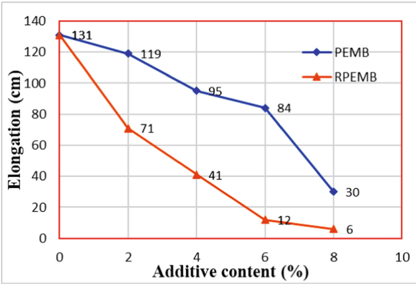

Fig. 8 Test result of elongation

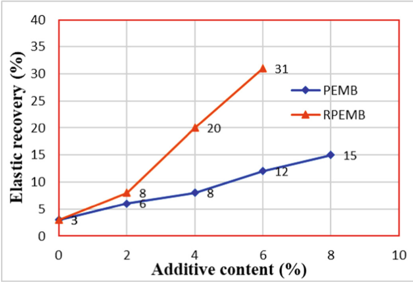

Fig. 9 Test result of elastic recovery

harder. The reduced elongation makes bitumen tend to decrease intrinsic strength, reduce intrinsic adhesion ability. With this test result, the additive content used should not more than 6%.

The elastic recovery of bitumen (deformation recovered after extending bitumen sample to 10 cm at 25 °C) increases from 3 to 6, 8, 12, 15% for PEMB and from 3 to 8, 20, 31% for RPEMB (Fig. 9), because of PE is good elasticity.

However, if the PE content is too high, the bitumen becomes stiffer and does not guarantee elongation. When RPE content is 8%, it makes the bitumen harder, the elongation is less than 10 cm, due to the composition of RPE (rough recycling) there are many hard plastics, many other impurities, not as homogeneous as PE. Improved elasticity recovery helps the bitumen reduce stiffness at low temperatures, and limits thermal cracking damage.

The adhesion to stone was also improved when using an additive content of 4% or more, it improved from level 3 to level 4, but the improvement was not so obvious through this testing (Fig. 10).

The viscosity of bitumen also increased significantly (Fig. 11). It increased more than 2.5 times (with PEMB) and nearly 2.0 times (with RPEMB) when the additive content reached 4%, increased more than 5.5 times (with PEMB) and nearly 3.0 times (with RPEMB) when the additive content is at 8%. The additive reduces the penetration, increases the softening point, so the increased viscosity is inevitable. The viscosity shows the properties of bitumen at a specified temperature. If the viscosity is too high, making the aggregate difficult to fully cover while mixing, the asphalt is harder, difficult for compacting. The viscosity is too low, making the aggregate and bitumen poor bond during construction, the AC is discrete, the material (in AC) is easy to peel. The suitable viscosity of bitumen is about 200 cp when mixed (at 150–160 °C), and about 2000–20.000 cp when compacting (110–135 °C), the viscosity of PMB is recommended not to more than 3000 cp at 135 °C. Test results show that viscosity of bitumen using PE/RPE improved at high temperature and still at a suitable level.

The storage stability of modified bitumen is shown by the difference in the softening point at 2 positions of the same sample: the upper and lower part of the standard sample tube (the sample after mixing is stabilized at 163 °C for 48 h). Storage stability shows dispersion and phase separation of PE in the PEMB colloidal system according to storage time. Test results in Table 4 show that PEMB has poor storage stability, phase separation occurs faster when PE content increases.

The storage stability depends on the PE content, resin and aromatic content in the bitumen composition, this is due to the mechanism of the PEMB colloidal system as analyzed in Sect. 2.1. The control of storage stability is very important for the PEMB colloidal system, which is also a fairly complex issue, which will be studied and presented in another paper.

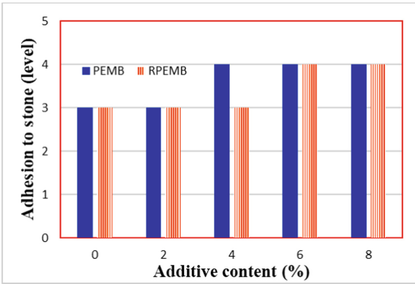

Fig. 10 Test result of adhesion

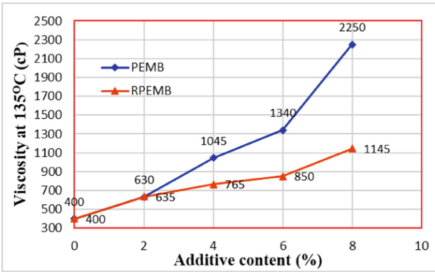

Fig. 11 Test result of viscosity

Table 4 Test result of storage stability

PE content (%)	2	4	6	8
	Softening point (°C)			
Upper part of sample	51.4	> 100	> 100	> 100
Lower part of sample	51.2	53.6	56.2	62.4

4 Conclusion

From the results and evaluation, it is initially shown that PE/RPE can be used as an additive like some other thermoplastic polymers to modify bitumen. They reduce the penetration, increase softening point, reduce elongation, increase elastic recovery and viscosity, make bitumen better bearing, improve resistance to deformation, rutting and abrasion, better anti-oxidant, more stable and less susceptible to high temperatures. However, they also make the modified bitumen tend to disperse, affecting the stability during storage. PE/RPE content should be used at 4–6%.

The properties of RPEMB have a different value than PEMB, this is because RPE (rough recycling) has many impurities, many hard plastic components, which have an adverse effect on the properties of bitumen.

It is necessary to continue to have researches on improving the storage stability of the PEMB colloidal system, the effect of impurities in the RPE on modified bitumen, the effect of the PEMB/RPEMB on the properties of AC. The research results will be presented in the next articles.

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