Improving Quality and Durability of Bitumen and Asphalt Concrete by Modification Using Recycled Polyethylene Based Polymer Composition

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Abstract

Bitumen is a component of asphalt binder that combines gravel, sand and mineral powder in the monolith. Bitumen performs this function due to adhesive properties and ability to become liquid when heated, and becomes solid when cooling. At the same thermoplasticity and low strength makes bitumen the most sensitive of all the components of asphalt to the effects of traffic loads and climatic factors. Softening of bitumen at summer temperatures causes rutting on roads and winter fragility results in the occurrence of low-temperature cracking. Modification of bitumen by such polymer additives as thermoplastics of SBS type, latex, terpolymers allows improving bitumen properties and increasing asphalt concrete durability. These additives increase cohesive strength and heat resistance of bitumen, impart elasticity to it, improve its behavior at low temperatures.

The main reason that hinders wide application of polymeric modifiers is their high cost. One promising way for solving an issue is partial substitution of expensive modifiers for cheaper plastic waste. In addition, this waste must meet two requirements. Firstly, it should be compatible with bitumen. Secondly, it must provide bitumen with the required positive properties, although maybe not to the extent as special modifiers do.

The conducted research showed that so-called recycled polyethylene resulting from the processing of plastic products (films designed for agricultural works, packaging material etc.) meets these requirements. Recycled polyethylene increases viscosity, cohesive strength and heat resistance of bitumen. The conducted studies allowed establishing effective combinations on the basis of recycled polyethylene which ensured significant reduction in the content of expensive special polymer modifiers in bitumen.
Ratio of components in the polymer composition is as follows: recycled polyethylene - from 50 % to 65 %, latex or SBS thermoeaplastics type - from 30 % to 50 %, plasticizer (industrial oils, oil extracts, etc.) up to 10 %. Rational content is from 3 % to 4 % by weight of bitumen. The technologies of combining polymer components among themselves and with bitumen were developed. Modification of bitumen by created polymer compositions improved strength and heat resistance characteristics of asphalt concrete and increased its resistance to cracking. Technological parameters of modified bitumen based asphalt mixtures preparation and placing were defined. Modification of asphalt concrete by direct introducing of composite polymer additive in the mix was proposed.

**Keywords:** recycled polyethylene; bitumen; asphalt concrete; modification

1 Introduction

The durability of asphalt concrete pavements is determined by the duration of their operation while preserving the required performance under traffic loads and weather and climatic factors. Bitumen produces a decisive impact on the durability of asphalt concrete which due to its bonding ability acts as binding component. When heated, bitumen becomes liquid allowing it to mix with a mixture of crushed stone, sand and filler. When cooled, bitumen becomes hard and turns mixture into a single monolith. However, thermoplastic properties of bitumen are one of the main reasons for the occurrence of deformations and deterioration on the pavement.

So, in summer bitumen softens at elevated temperatures and rutting is formed on the pavement under traffic loads. In winter bitumen becomes hard and brittle. Consequently, at low-temperature compression of asphalt bitumen cracks and transverse cracks occur on the pavement surface. Of all the components of asphalt bitumen is the least durable material and it has no elastic properties. Under the influence of long-term traffic loads stresses are accumulated in bitumen resulting in the occurrence of "fatigue" cracks on the pavement.

One of the most effective ways to increase the durability of asphalt pavements is modification of bitumen by special polymer additives, such as SBS type thermoeaplastics, latex, terpolymer and others. These additives increase heat resistance of bitumen, impart elasticity to it, improve behavior at low temperatures. Due to this resistance of asphalt pavements to traffic loads impact and adverse weather conditions increases and thus, their service life is prolonged.

The main reason that hinders a wide application of polymer modifiers is their high cost. One of promising ways for solving this issue is a partial replacement of expensive special modifiers with the cheaper general-purpose polymers or their waste. The main condition for this is imparting modified bitumen the required properties and compliance with current standards.

Among the general-purpose, high density polyethylene and recycled polyethylene – the product of recycling plastic waste polymers have the ability of combining with bitumen.

Given the above, the purpose of the study was to establish the possibility of using these materials as partial substitutes for such additives as latex, thermoeaplastics and terpolymers, and creating on their basis cheap and effective composite bitumen and asphalt modifier.

2 Research study

At the first stage of creating polymer modifying composition the effect of polyethylene and recycled polyethylene on the properties of bitumen was studied.
Then, to improve the efficiency of modification a special well-known modifier that, at the minimum contents could most effectively complement the main component of the polymer composition, was determined. Further an optimal ratio of components and the content of the entire polymer composition in bitumen were selected.

### 2.1 Materials and research methods

At the research fulfillment the following initial materials were used:
- Oxidized bitumen of Mozyr Oil Refinery production, brand BND 90/130.
- High pressure polyethylene (P).
- Recycled polyethylene in the form of white crumbs (RP).
- Anionic latex, brand Butonal NS 104 manufactured by BASF (Germany) which is the aqueous dispersion of copolymers of styrene (S) and butadiene (B) of irregular structure including conglomerated sulfur containing polymer phase (70 ± 1) % (SBR) (L).
- Thermoplastic elastomers of SBS type, Kraton D 1192 A grade of Kraton Polymers production (Netherlands) built from styrene block copolymer (S) - butadiene (B)- styrene (S) (TP).
- Terpolymer (reactive elastomeric thermoplastics polymer полімер) of Elvaloy AM grade of Dupont production (USA), which structure includes ethylene chains which the attached methyl acrylate functional groups and glycide methyl acrylate (TP).
- Plasticizing additive - industrial oil, brand I-40 (O).

Modification of bitumen comprises the following successive operations: heating of bitumen to a temperature (180 - 190) °C; gradual introduction of the desired quantity of polymer additives in bitumen and stirring it with the laboratory stirrer; mixing bitumen mixture with polymer over 2 hours.

Nomenclature of quality indices of source and modified bitumen and testing methods are shown in Table 1 (columns 1 and 11).

Standard content of thermoplastic elastomers or latex of SBS type in modified bitumen is 3 %, of terpolymer is 1.5 %. Given that, at this stage modified bitumen with the same content of polyethylene, recycled polyethylene and compositions based on them were investigated.

### 2.2 The research of polyethylene and recycled polyethylene impact on bitumen properties

Test results of initial bitumen and of bitumen modified by 3 % of polyethylene and recycled polyethylene, are shown in Table 1 (columns 2, 3, 4).

According to tests results, polyethylene can be combined with bitumen only partly and at the bottom of the reactor the sludge is left. At a low content (up to 1% wt) polyethylene hardly affects the binder’s properties, and at higher (3 % wt.) a considerable heterogeneity of binder (crumbs of undissolved polymer are available) takes place and significant deterioration in its structural and rheological characteristics is observed: sharply increasing viscosity and decreasing ductility.

Softening point of bitumen increases slightly (especially given the sharp increase in viscosity). Elasticity does not appear and fragility temperature slightly increases.

Compatibility of polyethylene with bitumen is somewhat improved with the introduction of a significant amount of plasticizer (30 %). Industrial oil, tar, oil extracts, etc. can be used as plasticizers. However, the presence of plasticizer does not significantly improve the properties of polyethylene modified bitumen.

Using recycled polyethylene - processed plastic products such as agricultural films, packaging materials, etc. turned to be much more promising.
During operation of production and processing of its waste polyethylene undergoes significant thermal and mechanical impacts resulting in partial destruction of the material, accompanied by local breaks of polymer chains and the growth of certain reactionary activity. Due to this, recycled polyethylene is much better than conditioned polyethylene combines with bitumen. It does not require the use of plasticizer and more effectively influences the binder’s properties.

When introducing 3% of recycled polyethylene, an increase in viscosity of bitumen (reduced penetration at 25 °C) in parallel with keeping its heat resistance high enough (which is manifested in higher softening point to 11 °C) is observed.

Recycled polythene does not impart elasticity to bitumen. Ductility of bitumen at a temperature of 25 °C drops sharply when recycled polyethylene is introduced. Presumably this is because the said polymer has significant deformability, probably of being in bitumen in the form of individual phases, it destroys homogeneity of the structure and thereby weakens ductility.

Ductility at 0 °C during modifications is also reduced but it is not worse than with the “clean” bitumen of similar viscosity. Brittleness temperature practically does not react to the presence of recycled polyethylene in the binder that can be considered as a positive thing given significant increase in the viscosity of bitumen.

The fulfilled tests demonstrated the possibility of using recycled polyethylene as the basis for effective polymer composition that is much cheaper than with foreign modifiers.

By tensile strength and elasticity bitumen containing recycled polyethylene does not meet the requirements of standard EN 14023.

2.3 Selection of modifying compositions based on recycled polyethylene

Further research has been focused on the selection and determination of the required amount of a well-known polymer modifier that with a minimum content would most effectively complement and enhance the effect of recycled polyethylene.

For this purpose, latex, thermoelastoplastics of SBS type and terpolymer were studied as an additional component.

To improve the combination of polymer components, both among themselves and with bitumen, plasticizer (industrial oil) was introduced in a polymer composition in some cases.
Bitumen containing the following ratio of the components of polymer composition was investigated:

- Recycled polyethylene/latex/plasticizer – 57/33/10 or correspondingly 1.7 %, 1.0 % and 0.3 % of bitumen mass (column 5 of Table 1).
- Recycled polyethylene/latex – 67/33 or correspondingly 2 % to 1 % of bitumen mass (column 6 of Table 1).
- Recycled polyethylene/latex – 50/50 or correspondingly 1.5 % to 1.5 % of bitumen mass (column 7 of Table 1).
- Recycled polyethylene/thermoelastoplastics/plasticizer – 50/40/10 or correspondingly 1.5 %, 1.2 % to 1.0 % of bitumen mass (column 8 of Table 1).
- Recycled polyethylene/thermoelastoplastics – 50/50 or correspondingly 1.5 % to 1.5 % of bitumen mass (column 9 of Table 1).
- Recycled polyethylene/terpolymer – 67/33 or correspondingly 2 % to 1 % of bitumen mass (column 10 of Table 1).

The results of testing bitumen with different content of polymer composition components are shown in Table 1 (columns 5 – 10 of Table 1).

Figure 1 presents the graph of bitumen quality indicators depending on composite additive content based on recycled polyethylene impact on bitumen properties.

The studies have shown that the introduction of 3% composition in bitumen keeping the following components ratio: recycled polyethylene - (50-65) %, latex - (30-50) % and plasticizer (industrial oil) - to 10 % allows obtaining modified bitumen conforming to the standard EN 14023 (columns 5, 6, 7 of Table 1).

It should be noted that the increase of latex improves characteristics of modified bitumen. At that, penetration decreases less rapidly at 25 °C, heat resistance of bitumen increases (increment of the softening point increases from 8 °C to 12 °C). Elasticity increases from 58 % to 66 %. Despite increased viscosity, brittleness temperature of modified bitumen does not change and ductility at 0 °C is 3 times increased. This indicates improvement in the behavior of bitumen at low temperatures.

When the content of latex in bitumen is less than 1 %, the introduction of plasticizer improves the properties of modified bitumen due to better compatibility with recycled polyethylene. At the content of latex of 1.2-1.5 % the plasticizer cannot be used due to the growing influence on the properties of bitumen and lesser dependence of binder characteristics on recycled polyethylene. The studies of
bitumen modified by 3% of polymer composition including, besides recycled polyethylene, SBS type thermoelastoplastics, were conducted (columns 8, 9 of Table 1). The content of the latter ranged from 30 % to 50 % of the composition volume or (1,0 – 1,5) % by weight of bitumen.

The study showed that at substitute of recycled polyethylene with termoelastoplastoplastics of SBS type bitumen viscosity increases less intensively than in case of using recycled polyethylene alone but more intensively than in case of using latex as an additional component. Increase in thermoelastoplastics content leads to higher softening point. When its content is increased from 1% to 1,5%, softening point value is increased by 5 °C. The expected decrease in ductility of bitumen at 25 °C was recorded and ductility grew by 30 % at 0 °C. Brittleness temperature remained unchanged. Elasticity of bitumen at 1,5 % of recycled polyethylene and 1,5 % of SBS type thermoelastoplastics content was 55 %. If you change the proportion towards the content of recycled polyethylene, elasticity will sharply decrease to 40 %. Analysis of test results showed that the use of thermoelastoplastics of SBS type modified bitumen has slightly worse physical and mechanical characteristics than bitumen modified by the composition in which the role of the second component latex was used.

To ensure the compliance with the standards requirements, minimum content of SBS type thermoelastoplastics in polymer composition must be at least 50 % or 1,5 % by weight of bitumen (at total content of polymer composition 3 %).

When using terpolymer as an additional component to the recycled polyethylene, negative results were obtained (column 10 of Table 1). Although the use of terpolymer provides modified bitumen with the highest softening point values, it fails to solve another major task - binder does not acquire sufficient elasticity and this indicator does not meet the requirements of EN 14023.

Along with the separate introduction of polymer components, bitumen modification can be performed using one composite polymer additive. In this case, the preparation of composite polymer additives is carried out by combining the polymer components (if necessary with plasticizer) using extruders or agglomerator where polymers are heated, fused and mixed, then cooled fused mass disintegrates in crumbs or granules.

The studies have shown that both at separate introduction of components and at preliminary their combining and use in the form of composite additive modified bitumen has almost identical properties. Designed composite polymer modifier was called Polydom.

### 2.4 Comparative analysis of composite additive based on recycled polyethylene and also other polymer modifiers impact on bitumen properties

Comparative studies of the properties of bitumen modified by composite polymer additive based on recycled polyethylene and the most known polymer modifiers: latex, thermoelastoplastics of SBS type and terpolymer were conducted.

At that, the content in bitumen of composition polymer additive Polydom, latex, thermoelastoplastics of SBS type was 2 %, 3 % and 4 %, and of terpolymer - 1,5 % and 2 %.

Test results are shown in Table 2.

References to the test methods are provided in Table 1, column 11.

Cohesion is defined using a cohesion test device. The device is developed by Kharkiv National Transport University (Ukraine).

Cohesion is a maximum stress at plane-parallel shear of two polymeric stripes between which bitumen sample is placed.

The studies have shown that at the introduction of 2 % Polydom fairly sharp drop in penetration of bitumen at 25 °C occurs and further increase in viscosity with increasing content of additives is observed.
Bitumen modified by other polymers is characterized by almost linear dependence of penetration reduction. Viscosity of binder increases rapidly with increasing terpolymer content but we must consider that the desired content of modifier is three times smaller than of other polymers. With optimal latex binder content the penetration is reduced by (20-30) 0,1 mm and for all other polymers - by (30-40) 0,1 mm.

Introduction of polymers significantly increases softening point of bitumen. Softening point increase with increasing Polydom content is linear and at 3 % it is up to 10 °C and at 4 % - 12 °C. By this indicator Polydom is almost identical to latex and yields to terpolymer and thermoelastoplastics of SBS type. Especially intensively softening point increases with increasing terpolymer content (if you add 1 %, softening point rises up to 14 °C, when adding 2 % - up to 21 °C, respectively). The increase of Polydom and other polymers content (except terpolymer) to 4 % has almost no effect on brittleness temperature. The stability of this index at increasing viscosity indicates of improving low-temperature behavior of modified binders. Thermoelastoplastics does not change brittleness temperature at the concentration to 2 %.

The tendency of brittleness temperature stability is typical for other modified bitumen. All polymers impart high elasticity to bitumen. At the introduction of the initial amount of polymer an intensive growth in the binder’s elasticity. Then, with increasing additives content stabilization of elasticity is observed. The highest elasticity (up to 80-88 %) has bitumen modified by terpolymer and thermoelastoplastics of SBS type. Polydom and latex have lower value (65-70 %).

Tensile strength at 25 °C at polymer introduction is expectedly reduced (in the case of Polydom up to 30-40 cm) and it remains stable at the modifier content from 3 % to 4 %.

Tensile strength at 0 °C for bitumen with Polydom is less than for bitumen modified by thermoelastoplastics of SBS type but it is higher than for "clean" indicating the preservation of higher plasticity by bitumen with Polydom at low temperatures compared to the initial bitumen.

When modifying bitumen by Polydom cohesive strength of bitumen is growing by more than twice. Among all polymers, Polydom bitumen provides with the highest cohesive strength. So, at 3 % content it makes 0,193 MPa. It is followed by terpolymer with the value of 0,184 MPa and then follow thermoelastoplastics of SBS type with 0,144 MPa and latex with 0,129 MPa respectively.

Table 2: Polydom in comparison with other polymer additives on bitumen of Lysychansk refinery production

<table>
<thead>
<tr>
<th>Indicators</th>
<th>Values of bitumen properties indicators with different content of polymer, %</th>
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</thead>
<tbody>
<tr>
<td>BND 90/130</td>
<td>terpolymer, %</td>
</tr>
<tr>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>1. Depth of needle penetration at temperature 25 °C, m×10⁻⁴ (0,1 mm)</td>
<td>100</td>
</tr>
<tr>
<td>2. Softening point (by ring and ball), °C</td>
<td>43,5</td>
</tr>
<tr>
<td>3. Ductility, m×10⁻⁵ (cm), at temperature 25 °C</td>
<td>&gt;100</td>
</tr>
<tr>
<td>4. Ductility, m×10⁻⁵ (cm), at temperature 0 °C</td>
<td>4,4</td>
</tr>
<tr>
<td>5. Elasticity, %</td>
<td>18</td>
</tr>
<tr>
<td>7. Cohesion, MPa</td>
<td>0,108</td>
</tr>
</tbody>
</table>
At the growth of Polydom content to 3 %, directly proportional dependence of cohesion growth is observed. At increasing Polydom content from 3 % to 4 %, cohesive strength growth is somewhat fading.

Comparative analysis of the results of testing Polydom modified bitumen showed the proximity of its properties to the properties of binders obtained using thermoelastoplastics and latex.

2.5 The research of composite additive impact on based on recycled polyethylene on asphalt concrete properties

The research of composite polymer modifier impact on asphalt concrete properties was conducted. Along with testing asphalt concrete based on bitumen modified by composite additive, the research of asphalt concrete modified by direct introduction of polymer additive in the mix during its preparation was also carried out.

For the research performance the following was prepared:
- Fine-grained dense asphalt concrete of type B containing 6 % of bitumen grade BND 90/130.
- Fine-grained dense asphalt polymer type B, which contains 6 % bitumen modified by 3 % polymer composition based on recycled polyethylene and latex - Polydom. Fine-grained dense asphalt concrete of type B containing 6 % of bitumen, brand BND 90/130 modified by direct introduction of 3 % and 4 % of recycled polyethylene and latex composition – Polydom.

Preparation of asphalt samples on the original and modified bitumen was performed in compliance with the standard procedure under DSTU B V.2.7-89.

Temperatures for heating materials at the preparation of the initial asphalt mix were as follows: for crushed stone, sand and filler - (165 - 170) °C, for bitumen - (135 - 140) °C. The mix was compacted at a temperature (145 - 150) °C.

When preparing asphalt mixes, modified bitumen, the temperature for heating materials were as follows: for crushed stone, sand and filler - (185 - 190) °C, for bitumen - (165 - 170) °C. The mix was compacted at a temperature (160 - 165) °C.

Preparation of asphalt mixes by direct introduction of rubber crumb to it included the following:
- Dosing and mixing of crushed stone, sand and filler followed by heating the mix to a temperature (185 - 190) °C;
- Adding the required amount of additives and stirring it with the mix;
- Adding of 6.0% bitumen heated to a temperature (140 - 145) °C to the mix and thorough mixing them until all the grains are enveloped;
- Forming asphalt specimens at temperature (160 - 165) °C under pressure of 300 kg / cm$^2$.

Test results are shown in Table 3.

<table>
<thead>
<tr>
<th>Indicators</th>
<th>Asphalt concrete</th>
<th>Physical and mechanical characteristics</th>
<th>Testing procedure</th>
</tr>
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<tbody>
<tr>
<td></td>
<td></td>
<td>polymer asphalt concrete containing polymer, %</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>In bitumen</td>
<td>In the mix</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3%</td>
<td>3%</td>
</tr>
<tr>
<td>1. Average density, kg/m$^3$</td>
<td>2,38</td>
<td>2,42</td>
<td>2,41</td>
</tr>
<tr>
<td>2. Water saturation, %</td>
<td>2,8</td>
<td>1,1</td>
<td>1,5</td>
</tr>
<tr>
<td>3. Swelling, %</td>
<td>0,35</td>
<td>0,2</td>
<td>0,21</td>
</tr>
<tr>
<td>4. Compression strength, MPa, at temperature:</td>
<td>3,8</td>
<td>5,8</td>
<td>5,3</td>
</tr>
<tr>
<td>20°C</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>50°C</td>
<td>1,6</td>
<td>2,8</td>
<td>2,5</td>
</tr>
<tr>
<td>0°C</td>
<td>10,2</td>
<td>9,3</td>
<td>9,4</td>
</tr>
<tr>
<td>5. Water resistance factor</td>
<td>0,91</td>
<td>1,0</td>
<td>0,98</td>
</tr>
<tr>
<td>6. Long-term water resistance factor</td>
<td>0,72</td>
<td>0,86</td>
<td>0,84</td>
</tr>
</tbody>
</table>

Table 3: Physical and mechanical properties of asphalt concrete modified by composite polymer
The studies have shown that when using modifier, strength and heat resistance of asphalt concrete is increased almost twice that ensures its high shearing resistance. In accordance with the obtained results, average density of modified asphalt concrete is higher than that of asphalt concrete based on source bitumen and it increases when modifier content grows, and water saturation, respectively, decreases.

Modified asphalt concrete displays the feature that is characteristic of all polymer asphalt concrete - lower temperature sensitivity. It has higher strength at high temperatures and lower strength at low temperatures. Temperature sensitivity factor defined by the ratio of asphalt concrete strength at 50 °C to its strength at 0 °C was increased on average by half that allowed predicting high heat and crack resistance of such polymer asphalt concrete.

Thus, the nature of changes of standard polymer asphalt concrete indicators based on Polydom is typical for polymer asphalt concrete obtained using commonly known polymer modifiers.

Performance of polymer asphalt concrete obtained by direct introduction of a modifier to the mix is slightly lower compared with the properties of asphalt based on modified bitumen but they also meet the requirements of Ukrainian standards DSTU B V.2.7-119.

To achieve the results identical to those that are achieved by adding 3% of bitumen modifier, 4% of composite polymer should be added in the mix.

Conclusions

- The possibility of reducing the cost of bitumen modified by partial replacement of expensive polymer modifier with cheap recycled polyethylene without impairing quality of binder is proved.
- Bitumen modified by polymer composition complies with the requirements of standard EN 14023 and has characteristics that are typical for using special polymer modifiers: latex and SBS type thermoelastoplastics.
- The best results were obtained when composite polymer modifier composition included latex (in the amount of 40 – 50 %) along with recycled polyethylene.
- Polymer asphalt concrete containing the designed modifier has higher strength, water and heat resistance values compared to conventional asphalt. It is also characterized by much lower temperature sensitivity and lower rate of thermal aging.
- The possibility of simplified technology of polymer asphalt concrete preparation by direct introduction of composite modifier in the mix during its preparation is proved.

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