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## POLYMER MODIFIED BITUMEN: REVIEW

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**Abstract.** The main reasons and ways of bitumen modification by polymers were examined. Positive and negative aspects of the usage of different polymer modifiers (elastomers, thermoplastic elastomers, reactive polymers, thermoplastics) were analysed. The authors' achievements concerning modification of bitumen by various polymers, namely indene-coumarone, petroleum, phenol-formaldehyde and epoxy resins, were described.

**Keywords:** polymer modified bitumen, elastomer, thermoplastic elastomer, reactive polymer, thermoplastic.

### 1. Introduction

According to [1] in 2013 ten major bitumen producers are: USA – 19.365 mln.tons (18.9 %); China – 17.938 (17.5); Russia – 6.193 (6.0); India – 6.193 (4.7); Canada – 4.335 (4.2); Iran – 3.896 (3.8); Japan – 3.659 (3.6); South Korea – 3.614 (3.5); Germany – 3.410 (3.3); Italy – 3.104 (3.0). These countries account for 68.5 % of world bitumen production. Ukraine occupies only 53 place – 0.123 mln.tons (0.1 %).

The largest exporters of bitumen are [2]: South Korea – 2.264 mln.tons (11.5 %); Iran – 2.179 (11.1); Singapore – 2.178 (11.1); Canada – 1.778 (9.0); USA – 1.539 (7.8). Due to the deficit Ukraine actually does not export bitumen.

The largest importers of bitumen are [3]: China – 3.225 mln.tons (19.8 %); USA – 1.641 (9.8); France – 1.193 (7.1); Saudi Arabia – 0.996 (5.9); Algeria – 0.729 (4.3). Ukraine imports 0.165 mln.tons (1.0 %; 21<sup>st</sup> place in the world).

World consumption of bitumen is on the average 87 mln.tons per year [5]. The structure of consumption by world regions is shown in Fig. 1.

About 85 % of petroleum bitumen are used as a binder in various types of asphalt laying: sidewalks, highways, airports, *etc.* [4, 5] (Fig. 1).

To date bitumen-mineral mixtures for pavement are prepared by two ways: (i) as a result of bitumen heating to the temperatures exceeding their dropping points; (ii) by obtaining aqueous bituminous emulsions. Table 1 represents the composition of petroleum bitumen consumption by the road industry in Europe according to EAPA (European Asphalt Pavement Association) [6].

When using bitumen for the production of asphalt mixtures it is not always possible to create a material that fully satisfied consumers by their performance properties. In particular, bitumen is generally characterized by poor adhesion, low temperature and plastic properties, leading to poor strength of the roadway. One of the ways of solving this problem is modification of commercial bitumen. Modified bitumen is characterized by improved flexibility and longer service life, it also has a lower brittleness temperature and higher softening temperature [7-10].

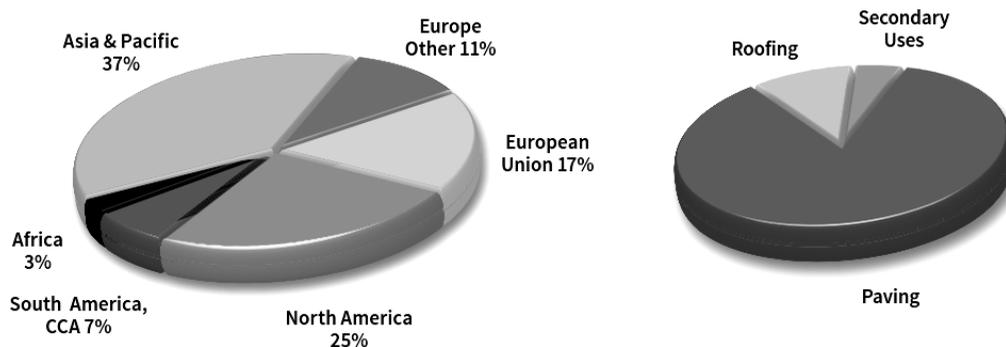


Fig. 1. World consumption of bitumen

Consumption of bitumen in the road industry [6]

Country	Tonnage in 2014, mln. t	Percent of modified bitumen (of total)				Emulsions in 2014, mln. t
		in 2011	in 2012	in 2013	in 2014	
Austria	0.37	25.0	25.0	30.0	18.9	0.005
Belgium	0.21	25.5	27.0	25.4	26.5	No data
Croatia	0.13	No data	No data	No data	10.00	No data
Czech Republic	0.33	18.0	22.4	22.1	21.5	0.008
Denmark	0.19	5.0	5.0	5.0	5.0	0.020
Estonia	0.08	No data	No data	4.0	3.3	0.016
Finland	0.27	0	No data	0	0	0.001
France	2.47	<10	13.0	13.0	6.9	0.792
Germany	1.60	No data	No data	No data	No data	No data
Great Britain	1.28	8.0	8.0	8.0	8.0	0.100
Greece	No data	2.0	1.5	2.2	No data	No data
Hungary	0.19	11.0	15.4	30.0	25.0	0
Iceland	No data	No data	No data	No data	0	0
Ireland	0.13	No data	No data	No data	No data	0
Italy	No data	14.0	13.0	14.0	No data	No data
Lithuania	No data	14.0	11.0	No data	No data	No data
Luxembourg	0.05	20.0	30.0	No data	15.0	No data
Netherland	0.30	No data	No data	5.0	No data	No data
Norway	0.35	15.0	No data	No data	No data	No data
Poland	No data	21.0	22.0	25.0	No data	No data
Romania	0.20	74.0	45.0	76.0	75.0	0
Serbia	0.26	14.7	6.0	4.2	4.4	No data
Slovakia	0.08	60.0	37.0	31.0	29.1	0
Slovenia	0.07	10.0	11.0	9.0	10.0	No data
Spain	0.56	13.1	19.6	10.0	15.0	0
Sweden	0.45	6.0	5.0	No data	7.0	0
Switzerland	0.28	10.0	11.0	2.0	12.0	0
Turkey	2.39	8.0	7.5	4.4	3.2	0.026

The share of modified bitumen, relative to its total use in road construction, is irregularly distributed between European countries, but as a rule, this share is above 10%. The leaders are (%): Romania – 75; Slovakia – 29.1; Belgium – 26.5; Hungary – 25; Czech – 21.5 (Table 1).

## 2. Bitumen Modification

As mentioned above, due to its natural properties petroleum bitumen is not able to create the conditions for long-term operation of pavement under modern heavy duty traffic and adverse weather factors.

Therefore, in addition to providing the required quality and durability it is necessary to radically improve the physical and mechanical characteristics of these materials by complex modification with additives.

From a technical point of view, to create a bitumen-based composite materials with a given set of properties only those substances may be applied, which meet the following requirements [17]:

- they are not destroyed at the temperature of asphalt-concrete mixture preparation;
- they are compatible with bitumen during the mixing process on conventional equipment at the temperatures usual for preparation of asphalt-concrete mixtures;
- in summer they increase the stability of bitumen (which is a part of the roadway) to the deformation impact loads without increasing its viscosity at the temperatures of mixing and laying and do not impart brittleness to bitumen at low temperatures;

- they are chemically and physically stable and do not change their properties during transport, storage, processing and operating conditions of pavement.

## 2.1. Classification of Petroleum Bitumen Modifiers

According to their action the modifiers can be divided into adhesion, plasticizing, structuring and complex ones.

Adhesion additives significantly improve bitumen adhesion to stone materials and, as a rule, bitumen particles with each other, provide high water resistance of asphalt-concrete and prevent damage of the pavement. Adhesion additives also retard the bitumen aging [11, 12].

Plasticizing additives provide modified bitumen with necessary consistency due to which they withstand deformation load and temperature changes [13].

Structuring additives can become cross-link binders and thereby provide the required strength of the roadway coating.

Complex additives are widely used in the road-building. They significantly improve the rheological properties of modified bitumen and their adhesion to mineral materials surface [14-17].

Additives for improving roadway quality are introduced into bitumen or directly into asphalt-concrete mixtures.

Additives which are introduced directly into bitumen can be divided into: polymer compounds, adhesive agents and specific components (*e.g.* sulfur). Additives introduced into asphalt-concrete mixtures are divided into: stabilizing (based on fibers), structuring (mineral powder, cement), structuring and stabilizing (natural bitumen, polymers, waxes). The compounds used to produce polymer modified bitumen (PMB) are described below.

Modification by polymer materials is one of the most perspective ways to improve the quality of binders to produce road surfaces with improved characteristics.

## 2.2. Polymer Modifiers

Polymer compounds, used as modifiers, may be assigned to one of four groups according to their nature, which implies a way to influence the properties of bitumen and method of introduction into PMB [7, 9, 16-17].

1) elastomers – natural and synthetic rubber, rubber crumb;

2) thermoplastic elastomers – block copolymers of butadiene and styrene type SBS;

3) thermosetting plastics – various copolymers and kooligomers (further – resins), which are usually have a large number of functional groups: epoxy, furfuro- and phenol-formaldehyde, carbamide, silicone, *etc.*

4) thermoplastics – polyvinyl acetate, polystyrene, polyisobutylene, polyethylene, polypropylene, atactic polypropylene, polyvinyl chloride, thermoplastics Elvaloy-4170 (copolymer of ethylene with butyl acrylate and glycidyl methacrylate), latexes of Butonal NS type, Viskoplast-S, ethylene methyl acrylate (EVA), petroleum resins.

By volumes of using the polymers are placed in the following order: thermoplastic elastomers of SBS type; thermoplastics of ethylene vinyl acetate type (EVA), polyisobutylene, polypropylene, a variety of block copolymers; polymer latexes and thermopolymers of ethylene glycidyl acrylate type [9, 10].

### 2.2.1. Elastomers

This type of modifiers includes polymers, rubbers and rubber polymers [9, 10, 18-24]. For the bitumen-polymer composition rubber imparts a new property – flexibility, which is typical of natural rubber in a wide temperature range. The new rheological state provides composition deformation at low temperatures, despite the lack of plastic properties of bitumen. In other words, this additive in bitumen improves its extensibility and elasticity at low temperatures, improves thermal and crack resistance, strength, deformation resistance, water resistance, frost resistance, durability, resistance to aging.

Rubber can be introduced into bitumen by direct fusion, or in the presence of a solvent.

The example of most suitable for bitumen modification elastomers is synthetic rubbers of general purpose (isoprene, divinyl, divinyl styrene, ethylene propylene). These rubbers are produced as a briquetted solid elastic product which is used for gum production *via* rubber vulcanization.

The main disadvantage of elastomers is their poor dispersion in bitumen, so special measures are needed to obtain homogeneous modified bitumen (fusing, special solvents, *etc.*). Moreover, phase separation between polymer and bitumen in a liquid phase takes place, especially during storage. To avoid this phenomenon complicated devices that can support high temperatures and PMB continuous mixing are needed.

According to the published data [23, 24] rubber crumb is a promising product to be used for the modification. The main advantages are low cost and possibility of tires and other waste rubber products recycling. Despite the obvious advantages over other modifiers, using crumb rubber in road building is also limited because of the technological difficulties that arise when they are mixed with bitumen.

### 2.2.2. Thermoplastic elastomers

There are three types of styrene block copolymers: styrene-butadiene-styrene (SBS), styrene-isoprene-styrene (SIS), styrene-ethylene / butylene-styrene (SEBS). The

content of polymers in PMB can reach 3–10 wt %. Thermoplastic elastomers (TE), compared to thermoplastics, are characterized by higher elasticity, *i.e.* TE combine the strength of thermoplastics and elasticity of elastomers [7, 9, 10, 18-20, 25-27].

Styrene block copolymers of SBS type are mostly used for bitumen modification due to their ability to increase the strength of bitumen, and to provide polymer-bitumen compositions with elasticity, including that at low temperatures. Thermoplastic elastomer of SBS type is a linear polymer, which provides elasticity (ability to initial distortion) for bitumen-polymer system due to its structure (styrene units are linked by butadiene "flexible" blocks). TE retain the ability to highly elastic deformations within the temperature range of 193–353 K. The temperature of TE destruction is 463–483 K.

Block copolymers of butadiene and styrene of SBS type are well combine with bitumen because polystyrene and polybutadiene are swelled in paraffin-naphthenic and aromatic hydrocarbons of bitumen and partially dissolved in them at 423 K.

To improve polymer dissolution in bitumen the plasticizers are used (usually a mixture of aromatic based hydrocarbons). Plasticizer significantly improves the polymer solubility in bitumen, but slightly decreases the adhesive properties of the resulting binder compared to the original bitumen [7, 10].

Due to their properties TE are produced in a variety of trademarks [28-31]: Kraton D1101, Kraton D1184, Kraton D1186 (Kraton Polimers, USA); Finaprene 411 (Total, France); Europrene Sol T 161B (Eni, Italy); Calprene 411 (Dynasol, Spain); DST 30-01 and DST-30R-01 (Voronezh factory, Russia).

The main disadvantage, restraining the growth of modified bitumen production is its high cost. Bitumen modified by TE is 1.5–2.5 times more expensive than unmodified bitumen [8].

### 2.2.3. Thermosetting plastics

Thermosetting plastics (TP) are polymers, which turn into solid state while heating or introducing hardener. Before hardening TP molecules have a linear structure, the same as thermoplastics molecules, but the size of their molecules is significantly smaller. TP molecules are chemically active. They contain either double (unsaturated) bonds or chemically active groups. Therefore, under certain conditions (at heating, irradiation or adding hardeners) thermosetting molecules react with each other and form a continuous network. Epoxy, phenol-formaldehyde, carbamide, polyester, silicone and other resins belong to TP [7, 9, 10, 32-36].

Bitumen-polymer binders with thermosetting plastics have relatively high adhesion to the mineral particles and high strength.

The use of TP for bitumen-polymer mixtures create a number of disadvantages [7]:

- when entering hardener the technological properties of PMB are almost immediately deteriorated;
- rigidity is increased at low temperatures;
- the use of special hardeners complicates the system and raises its price;
- the effectiveness of thermosetting plastics appears usually at their large quantities in bitumen – more than 10 wt %.

### 2.2.4. Thermoplastics

These polymer additives improve strength and deformation characteristics of bitumen and asphalt: the resistance to rutting at elevated temperatures increases, risk of cracking at low temperatures and fatigue cracking of asphalt under prolonged stress is reduced. Polymers which are quite often used now: polyvinyl acetate, polystyrene, polyisobutylene, polyethylene, polypropylene, atactic polypropylene, polypropylene, polyvinyl chloride, Viskoplast-S, EVA, ethylene methyl acrylate [7, 9, 10, 18, 20, 37-40]. However, the use of "traditional" polymers (polyethylene, polypropylene, *etc.*) is accompanied by a number of problems because rigidity and brittleness of resulting PMB increase after modification. As a result, a pavement using such binders, is disposed to increased cracking at low temperatures.

Synthetic latex of Butonal NS type (BASF, Germany) [41-44] and thermopolymers of Elvaloy series (DuPont, USA) [7, 18, 45] are widely used. These polymers not only reduce bitumen sensitivity to temperature changes, but also enhance the cohesive and adhesive strength resulting in the increase of strength and crack resistance of asphalt concrete pavement.

Petroleum resins obtained from by-products of fossil fuels thermodestruction occupy a special position among thermoplastics. They can be used to improve bitumen properties, namely they increase the viscosity, hardness, durability and adhesion to mineral materials [46- 48]. The additives containing polar groups in their molecules, contribute to improved wettability of the mineral materials with bitumen, creating an adsorption and hemisorption monomolecular layers providing strong adhesion of bitumen-mineral mixture.

Petroleum resins with functional groups are of special attention. These compounds may belong both to thermoplastics and thermosetting plastics. The presence of functional groups in petroleum resin structure significantly improves the adhesion properties of bituminous materials, resulting in increased durability of the products based on them. The use of petroleum resins for bitumen modification is limited due to their considerable cost. Moreover, under high temperatures these compounds can form three-dimensional cross-linked

structure with petroleum bitumen that reduces its plasticity. As a result cracking of asphalt pavement occurs. Under long-term exposure to high temperatures during preparation, transportation and laying of road surfaces thermal degradation of polymers is also possible, so they can lose modifying properties.

Resin modifiers may also be produced from coal processing secondary products. The investigations concerning using indene-coumarone resin (ISR) as a modifier were carried out at the Department of Oil and Gas Processing of Lviv Polytechnic National University [49-51]. ISR is a polymerization product of a mixture of compounds (mainly indene, coumarone and styrene), which are by-products of the coking process ("heavy" benzene, coal tar light fractions). ISR introduction into bitumen significantly increases its softening temperature (from 320 to 325 K) and significantly improves the adhesion properties (Table 2). On the other hand, the plastic properties of bitumen are deteriorated (penetration and ductility decrease), the same as while using other thermoplastics and majority of thermosetting plastics. Therefore, as a plasticizer it is proposed to use tar and/or resin, which is a by-product of brown coal oxidative desulphurisation [52-57].

### 2.3. Non-Polymer Modifiers

Other types of modifiers (non-polymer modifiers) should be mentioned here, as well. Sulfur is considered to be the most promising among them. Technical sulfur is inexpensive and small-scale product. While producing sulphur asphalt the part of expensive bitumen is substituted for relatively cheap sulfur (50 wt %). The result is reduction of the finished product price and improvement of asphalt properties. Sulfur asphalt has several unique properties such as high compressive and flexural strength, adhesion, chemical resistance, frost resistance, low water absorption and waterproofness. A major disadvantage of sulfur compositions is their low thermal stability. In addition, there is a significant change in volume during sulfur solidification due to phase transition of sulfur from a liquid to a solid state [58-61].

Mineral materials are often added to bitumen but they should be rather fillers than modifiers. For example,

the wastes obtained during haydite and soda production are added to bitumen. The use of haydite dust improves crack resistance and strength characteristics of the bituminous binder. When using wastes of soda production the asphalt strength is quite high but water resistance after long-term water saturation is below limit. This problem may be solved by using silica powder [62]. However, mineral fillers do not usually improve adhesion and/or plasticity of bitumen.

### 3. Conclusions

85 % of bitumen production (on the average about 90 mln.tons of bitumen per year) is used as a binder in road construction. However oxidized and residual bitumen does not have the necessary properties to meet modern requirements to the pavement.

The most common way to increase the performance properties of oil bitumen (primarily, oxidized bitumen) is the use of various polymers as modifiers. Polymer modified bitumen compared to conventional bitumen has a number of significant advantages. They are more elastic, flexible and durable, less sensitive to the temperature changes and aging, have better adhesive and cohesive properties.

Thermoplastic elastomers, particularly SBS, are most widely used as bitumen modifiers due to their complex action. They provide bitumen with high strength (the same as thermoplastics and thermosetting plastics), elasticity (as elastomers), heat resistance, adhesion, and improved low-temperature characteristics. But the main reason that restrains thermoplastic elastomers prevalence is their high cost. An effective solution of this problem is their partial or complete replacement by cheaper polymers.

A promising direction is the production of modifiers from by-products of fossil fuels thermo-destruction, which contain a sufficiently large amount of unsaturated compounds. Among mentioned by-products are "heavy" benzene and/or coal tar light fractions, obtained at coke plants and used for the production of indene-coumarone resin. The latter one can be used to produce modified bitumen, which meets regulatory requirements and has extremely high adhesion (till 100 %).

Table 2

ICR modified bitumen

Plasticizer	Blend composition, wt %			Main characteristics of resulting PMB			
	bitumen	ICR	plasticizer	Softening temperature (ball & ring method), K	Ductility at 298 K, m·10 <sup>-2</sup>	Penetration at 298 K, m·10 <sup>-4</sup>	Adhesion to glass, %
–	100.0	0	0	320	75	62	46
–	93.0	7.0	0	325	36	38	100
Resin of brown coal thermodestruction	84.0	7.0	9.0	325	28	62	100
Tar produced from West-Ukrainian oils [52-57]	85.0	7.0	8.0	325	26	60	93

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## ПОЛІМЕР-МОДИФІКОВАНІ БІТУМИ: ОГЛЯД

**Анотація.** У статті розглянуті основні причини та способи модифікації бітумів полімерами. Проаналізовано позитивні і негативні сторони використання різних груп полімерних модифікаторів (еластомери, термопластичні еластомери, реакційні полімери, термопласти). Описано досягнення авторів статті в області модифікації бітумів полімерами: інден-кумароновою, нафтополімерними, фенолформальдегідними і епоксидними смолами.

**Ключові слова:** полімер-модифікований бітум, еластомеи, термопластичні еластомери, реакційні полімери, термопласти.