



TECHNOLOGY OF PREPARATION OF THE WATERPROOFING MATERIAL ON THE BASIS OF ASPHALT-RESIN-PARAFFIN DEPOSITS AND POLYMER

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ABSTRACT

In this paper described ways of recycling asphalt resin paraffin deposits (ARPD). Processing of oil waste and reducing their formation is a significant problem from the ecological point of view and requires new ecological and technological solutions. In this research paper analyzes the results of world practical experiments on the application of waste and presents technical solutions for the use of waste that are suitable for future application. The feature of this work is obtaining waterproofing materials based on asphalt - resin - paraffin deposits and polymer, moreover the improvement of its physicochemical properties.

Keywords: asphalt - resin - paraffin deposits, polymer, waterproofing material, oil wastes, polymerorganic material.

INTRODUCTION

The problem of environmental safety in the handling of solid oil waste is relevant worldwide, but is particularly acute in Kazakhstan nearly every oil-producing region.

As our studies have shown, handling of oil wastes should include the development of economically accessible and technically feasible technologies for involving waste in the resource flow. Requires new methodological approaches to solve the problem of disposal of oil waste not traditional destructive methods and techniques to improve consumer properties, removal of unwanted impurities and components, concentration, dehydration and other enrichment methods with the use of waste in the related areas of production. Such approaches to the involvement of waste in the resource turnover should be the basis of the strategy for handling oil wastes and related technical solutions.

The oil production management system should include the following stages: formation, separate accumulation and collection, transportation, processing, neutralization and placement in the environment of non-recoverable residues. In the current practice, the handling of oil wastes is reduced to their joint collection, transportation and temporary placement of qualitatively different waste streams, which makes it difficult to continue using them.

One of the ways to solve the emerging environmental and economic problems is the use of ARPD in the production of waterproofing building materials, for example, to obtain polymerorganic covering compositions.

Traditional materials for creating waterproofing systems are oil bitumen. The valuable properties of bitumen, which caused their widespread use in construction, are resistance to acids, alkalis and other aggressive reagents. At the same time, under the atmospheric influences, bitumens age, and their protective properties deteriorate. In order to strengthen the structure and improve the mechanical strength of bitumen, its

combination with related materials of organic origin is widely practiced.

Improving the quality of bitumen materials is achieved by modifying them with polymers using natural and synthetic rubbers, resins, ethers, etc. (Stabnikov, *et al.*, 2005). In the process of fusing with the polymer bitumen in the bitumen-polymer system implementing a mutual diffusion of these materials. Bitumen molecules with a high mobility penetrate the polymer structure, as far as their size and free space between polymer macromolecules allow. In this case, and separate groups of polymer molecules diffuse into bitumen. The result of this interdiffusion occurs coagulation of bitumen-polymer structure that can perceive the deformation over a wider range (Stabnikov, *et al.*, 2005).

By varying the component composition of the polymer-bitumen compositions, it is possible to obtain materials with predetermined physico-mechanical and chemical properties.

Thus, the analysis of previously published studies confirmed the possibility of using oil waste and polyethylene waste in the compositions of polymer-bitumen compositions to produce high-quality products with specified properties.

The study of the composition and properties of ARPD showed that the deposits are hydrophobic, have affinity for structure and high adhesion to synthetic thermoplastics such as polyethylene. This determines the possibility of utilizing the deposits in waterproofing polymerorganic material, inexpensive and easy to manufacture, based on secondary materials.

EXPERIMENTAL PART

Considering the fact that ARPD and bitumen analogues in the component composition, it can be assumed that the use of deposits will achieve high preset properties of the material at lower costs.

Since the predominant components in the ARPD are high-molecular organic compounds, this allows us to conditionally refer ARPD to organic materials. And in



order to emphasize the difference in the already developed organomineral waterproofing material on the basis of ARPD and the existing polymer-bitumen materials from the developed composite material based on ARPD and polyethylene, we conventionally called this material "polymerorganic".

The choice of components for the creation of an polymerorganic material was carried out taking into account their availability and the availability of certain

physicochemical properties, their possible compatibility in the composition with the obtaining of qualitatively new properties different from each component separately.

Analysis of the results of the known technical solutions in the field of polymer-bitumen compositions, presented in Table-1, made it possible to propose a hypothesis about the expected composition of the polymerorganic material, using the following: weight, %: ARPD-35 ... 90, polyethylene waste - 10 ... 65.

Table-1. The composition of the polymer organic material.

Component	Composition, weight %			
	Analogue 1 [4]	Analogue2 [5]	Analogue 3 [7]	Developed composition
Bitumen	33-50	90	25-33	ARPD
Plasticizer (oilsludge)	—	—	30-50	35-90
Filler	—	—	20-25	—
Polymerplast	50-67	10	5-12	10-65

When developing a material with specified properties, it is necessary to establish significant factors that determine the processes of structure formation in the "ARPD - thermoplast" system. These factors include: the chemical composition and properties of the ARPD; Type of polyethylene (depends on the method of production) and, accordingly, the physicochemical properties of

polyethylene; General and environmental requirements for the material (Tanzharikov, *et al.*, 2015). To determine the most significant factors, it is necessary to analyze the entire system. Interrelations and mutual influence of the factors determining the formation of the structure of the polymerorganic material using ARPD and polyethylene are shown in Figure-1.

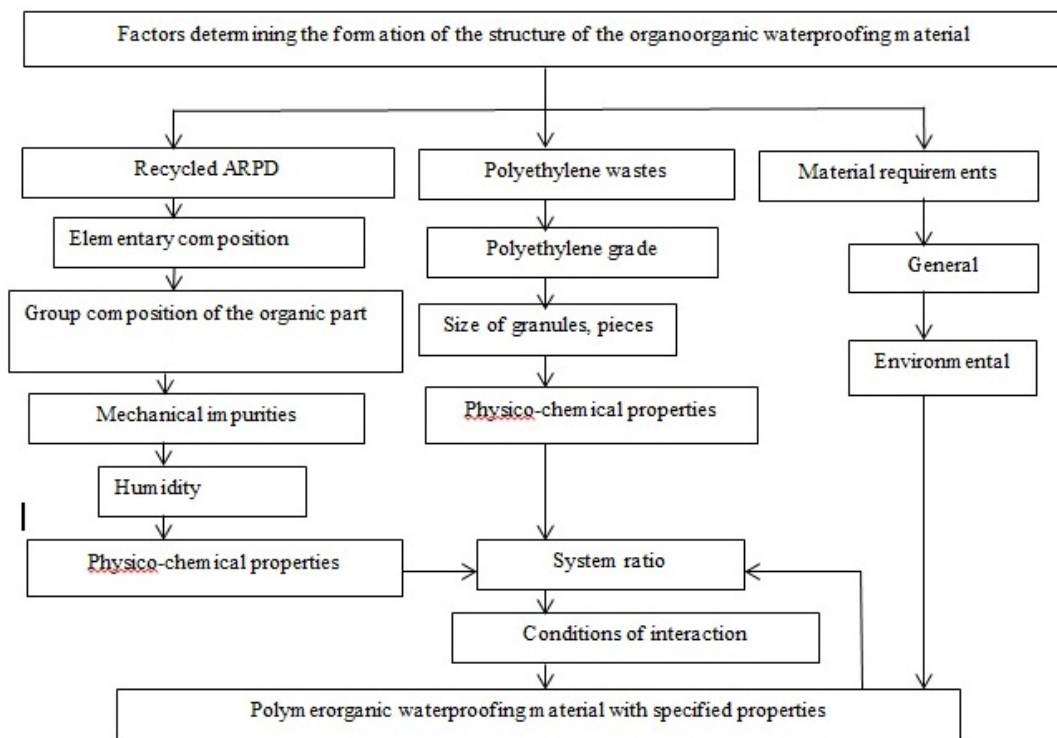


Figure-1. Analysis of the formation process of the structure of polymerorganic waterproofing material.

Analysis and evaluation system of the factors determining the process of forming the structure of the

developed material, it possible to identify the main controllable parameter - the ratio of components in the



system "ARPD - Thermoplastic" and the control parameters such as physical and mechanical properties of the material. The specified properties are determined by the requirements (technical and environmental), established by regulatory documents to the material. Polyethylene has the ability to soften when heated, and solidify upon cooling, that is, it is a thermoplastic. Polyethylene is produced in installations of high (150-300 MPa), medium (3-4 MPa) and low (0.25-0.5 MPa) pressure. The preparation process significantly affects the properties of polyethylene (Table-2) (GOST 11506-73).

High-pressure polyethylene differs branched structure, the number of branches is from 20 to 40 per 1000 carbon atoms and branches may have a different length, but basically it is a chain with carbon atoms 2 and 4. Macromolecules of low-pressure polyethylene have significantly fewer lateral branches, than high-pressure polyethylene. Basically, these are metal groups; there are no long-chain branches. There are only 5-15 side branches per 1000 molecules of the polymer, and this is mainly the CH_3 group. Medium pressure polyethylene is a practically linear polymer. The proportion of branching in it is insignificant (1.5-5 per 1000 atoms of carbon).

Table-2. Properties of polyethylene.

Properties	polyethylene		
	high-pressure polyethylene	low-pressure polyethylene	Medium- pressure polyethylene
Molecularweight, thousandunits.	2-40	70-3000	70-400
Degreeofcrystallinity,%	53-67	80-90	85-93
The number of CH_2 groups per 100 carbon atoms (branching)	21,6	5,0	1,5
Density, kg / m	917-930	940-960	960-970
Meltingpoint, °C	105-108	120-125	127-130
Heatresistance, °C	108-110	120-128	128-133
Breakingstrength, MPa	11-16	20-35	27-33
Flexuralstrength, MPa	10-17	20-38	2540
Brinellhardness, MPa	1,4-2,5	4,5-6,0	5,6-6,5
Relativeelongationatbreak,%	400-600	200-900	
Waterabsorptionfor 24 hours,%	0,01	0,005-0,032	0,01-0,02
Operatingtemperaturerange, °C	-70+110	-70+130	-70+130
Dielectric loss tangent at 10^8 Hz	2-10 ⁻⁴	2-10 ⁻⁴	2-10 ⁻⁴
Specificvolumetricalelectrical Resistance, Ohm*cm	10 ¹⁷	10 ¹⁷	10 ¹⁷

The features of the molecular structure (mainly branching) associated with the polymerization conditions determine the main technological properties of certain types of polyethylene. Branching, especially long-chain hinders the dense packing of macromolecules and reduces the crystallinity of the polymer. The most branched high-pressure polyethylene is a soft and elastic material, relatively little crystalline, has a lower density and a melting point. Medium-density polyethylene, in contrast, has a maximum degree of crystallinity, the highest density, and a higher melting point. Polyethylenes of medium and low pressure are rigid materials.

Film polymeric materials have good physicomechanical and anticorrosion properties, of which for hydraulic engineering are the most significant such as strength, density, hardness, water resistance, frost resistance, long-term strength, chemical resistance, weather resistance.

The water absorption of dense hydrophobic polymer materials is 0.005-0.02%. Due to high non-

permeability, polymer film, roll and mastic materials, especially based on polyethylene, have found application for waterproofing (Uliya, *et al.*, 2016).

By polyethylene disadvantages are, first of all, low oxidation resistance, and thermal and photoaging. To prevent this, thermal and light stabilizers are used. For resistance to thermal oxidative degradation, light and atmospheric aging, high-pressure polyethylene surpasses low-pressure polyethylene.

Improving the properties of the polymer can be achieved by their modification with acids, natural resins and bitumen, low-molecular rubbers and other substances. Modification is carried out in the process of obtaining a polymer by replacing some functional groups by others, grafting blocks of one polymer to the main chain of another (Uliya, *et al.*, 2016).

High physical and mechanical properties of polyethylene, the ability to improve the properties of bituminous materials due to their compatibility in polymer-bitumen compositions determine the possibility



of using polyethylene to create anpolymerorganic composition based on ARPD.

Given the characteristics of the molecular structure defining basic technological properties of various types of polyethylene, in the present study investigated the influence of the type of polyethylene (high and low pressure) on the physicommechanical properties of the polymerorganic material using ARPD.

The use of polyethylene waste for the manufacture of waterproofing coating will eliminate the use of commercial polymer and expand the scope of this secondary product. Most chemical compounds of deposits at low temperatures are water resistant and inactive, which makes it possible to use ARPD in waterproofing materials that are resistant to solutions of weak acids and alkalis. The presence of a significant amount of paraffins indicates good anti-corrosion properties of ARPD, which can occur in materials for a long time.

The high concentration of natural surfactants in ARPD provides strong adhesion to the materials. Therefore, from the chemical point of view, the improvement of ARPD is practically not required. The possibility of using ARPD for waterproofing purposes will be determined by the requirements for waterproofing materials. On the one hand, binders must have a low viscosity to ensure the preparation of a high-quality waterproofing composition at the lowest temperature, Method and a uniform thickness of the coating. This requirement of ARPD is fully met. On the other hand; waterproofing binders must be resistant against aging under the influence of weather and climate factors, heat-resistant, deformable in a wide range of temperatures. Strength and deformation properties of ARPD are determined by their physical structure - the ratio of structural elements: hydrocarbons, resins, asphaltenes. So the low value of the ratio of asphaltenes to the sum of hydrocarbons and resins 0.01 - 0.07 indicates a low melting, ductility and low strength of deposits. Therefore, in this case ARPD can not be used in waterproofing materials without adequate structure improvement.

One of the ways to improve the deformation and strength properties of ARPD can be their combination with polymer thermoplastic additives. It is known that the use of thermoplastic elastomers without fillers in combination with bitumens is the most technological additive, because when heated they melted and, with mixing, form a homogeneous mixture in a very short time (Tanzharikov, *et al.*, 2015).

Consequently, based on the studies carried out on the fusion of bitumen with thermoplastics of linear structure, it can be assumed that the combination of ARPD with polyethylene-type thermoplastics will result in a structural improvement in ARPD. The main role in the formation of a new structure of the organic polymer

material based on the ARPD-polyethylene system should belong to the physico-chemical transformations of the ARPD with the formation of a spatial lattice of a high polymer that plays the role of an elastic reinforcement that prevents the aging of deposits in the case of their heating and maintains equilibrium in the ARPD due to the return of some hydrocarbons.

To confirm the hypothesis put forward, experimental studies were carried out to develop the optimal composition of the polymerorganic material based on ARPD and polyethylene waste and to study the microstructure of the material. Analysis of the formation processes of the polymer organic waterproofing material has shown that the production of a material with specified properties that meet the requirements of the waterproofing structure to the material is determined by a combination of factors: the composition and properties of the ingredients, their quantitative ratio, mechanical and physico-mechanical effects.

The field of application of waterproofing materials is determined by the specific operation of various engineering structures. Our research was aimed at the development of organic polymer compounds based on ARPD and polyethylene waste, which can be used, firstly, for anti-filtration screens (given the great need for inexpensive and affordable waterproofing materials), secondly, as roofing and waterproofing mastics.

The composition of the polymer organic waterproofing material for anti-filtration protection of waste landfills should have elastic properties, low water absorption and a filtration coefficient, high resistance to aggressive media (Shomantayev, *et al.*, 2016).

Based on the engineering calculation of the waste storage sites, the strength of the material is most fully characterized by the compressive strength and water absorption limit. Waterproofing material during operation should not have a negative impact on groundwater.

Therefore, based on the established technical and environmental requirements for anti-filtration protection, the following parameters were adopted as optimization parameters for the composition of the polymer organic waterproofing material being developed: compressive strength; Water absorption; Coefficient of filtration.

To justify the construction of the anti-filtration screen using the material being developed, extraction of petroleum products was determined upon contact with water. As a result of optimization, we sought to obtain a material with maximum compressive strength and minimum water absorption.

The composition of the polymerorganic material based on ARPD and polyethylene waste for roofing and waterproofing mastics must meet the general requirements for mastics in accordance with (GOST 30693-2000) (Table-3).

**Table-3.** General requirements for mastics.

The name of the indicator	Unit of measurement	Bitumen-polymer mastic (hot)	
		For gluing bitumen and bitumen-polymer roll materials	For mastic roofing or waterproofing
Conditional strength,	MPa	—	0,2
no less			
Relative elongation at break, no less	%	—	100
Strength of adhesion to the base, not less than	MPa	0,1	0,1
Strength of adhesion between layers, not less than	MPa	0,1	—
Shear strength of adhesive compound, not less than	kN/m	0,1	—
Water absorption (by weight), no more than	%	—	2
Flexibility on a bar with a radius of 5.0 mm, not less than	°C	-15	- 15

RESULTS AND DISCUSSIONS

Thus, the composition of the polymerorganic material, which is applicable as roofing and waterproofing mastic for the roofing, must have a conditional strength of at least 0.2 MPa, a relative elongation at break of at least 100%, shear strength with a base of not less than 0.1 MPa and water absorption is not more than 2%. In the experimental studies, the quantitative composition of the components was the main variable factor determining the achievement of the given properties of the material. To determine the optimal compositions of polymerorganic waterproofing material, we conducted laboratory studies of samples of various combinations of composition. The main component of the polymerorganic material is ARPD. In the experimental studies, the ARPD of the Kumkol field was used. The polymer used high pressure polyethylene (HPPE), grade 10803-020 (GOST 16337-77) in the form of granules with a diameter of 5 mm and waste film of 0.2 mm thickness, and a low-pressure polyethylene (LPPE) grade 19013-002 (TU 6 -05-1853-78) in the form of granules with a diameter of 5 mm.

In order to achieve the greatest effect when combining ARPD and polymer and obtaining a material with predetermined properties, a set of optimal mechanical and physicomachanical effects was applied by heat treatment of the mixture with vigorous stirring (Bisenov, *et al.*, 2016). Mechanical stirring was carried out in a propeller-type mixer at a speed of 1400 rpm for 45

minutes. Stirring was carried out at a temperature of 130 °C. The obtained polymerorganic material was cooled in air and, after 3 hours at room temperature, its physicomachanical properties were determined. Studies of the physico-mechanical properties of polymerorganic compositions included the definition of: compressive strength; appearance, strength of adhesion to the base, flexibility, conventional strength, extensibility, relative elongation and water absorption; softening point; depth of penetration of the needle (penetration); temperature of brittleness, filtration coefficient. In addition, extraction of petroleum products in an aqueous solution from samples of a material was determined. The concentration of petroleum products in the aqueous solution was determined by gravimetric method, after contact for four weeks of a sample weighing 100 g in a sealed container with water of 1 dm³ (mixture / water ratio 1:10).

Dispersion analysis was used to describe the data and to assess the statistical significance of the results of the studies.

The first stage of laboratory studies included the manufacture and testing of samples of polymerorganic material with a wide range of variations in the mass content of the components in the formulations, %: ARPD 35-90, polyethylene waste 10-65, based on an analysis of similar technical solutions (see Table-1). The compositions of the investigated samples of the material are given in Table-4.

**Table-4.** The compositions of the investigated samples.

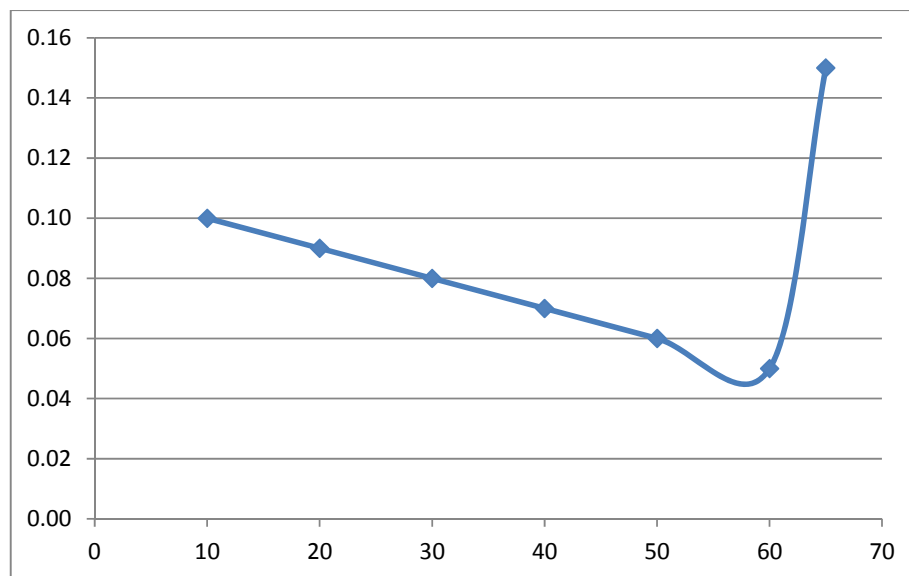
Components	Mass content of components in the composition, %						
	1	2	3	4	5	6	7
HPPE (granules) LPPE (granules) HPPE (film waste)	10	20	30	40	50	60	65
	10	20	30	40	50	60	65
	10	20	30	40	50	60	65
ARPD	90	80	70	60	50	40	35

As a result of the studies, it was found that with the increase in the composition of the polymer organic material with polymer from 10 to 60%, the strength of the material increases. Moreover, if the compressive strength of the samples obtained by adding 10% by weight of the polymer is minimized, and the strength of the samples with a content of 20% or more in the polyethylene composition is compared with this value, it turns out that the addition of 20-40% of the polymer leads to an increase Strength in 1,7-2,2 times for compositions in HPPE (film), in 1,9-2,6 times for compositions with HPPE (granules) and in 1,8-2,4 times for compositions with LPPE. The addition of 50-60% polyethylene in ARPD increases the strength of polymerorganic material by 3.4-4.6 times for compositions with HPPE (film), 4.2-6.3 times for compositions with HPPE (granules), in 3.2 -4.3 times for compositions with LPPE. Increased mass polyethylene content of more than 60% of the composition made it

difficult method of preparation due to insufficient amount of binder and degrades the deformation properties of the resulting material.

The obtained data indicate that the formulations have a maximum strength to weight content: polyethylene waste (regardless of brand) 50-60% and 40-60% ARPD. It is known that the increase of polymer in polymer-bitumen compositions reduces the hydrophilicity of the material and, correspondingly, decreases the water absorption [1]. A similar conclusion is also confirmed with respect to the reduction of the filtration coefficient of the proposed polymerorganic material with an increase in its polymer content.

To confirm the literature data with reference to the ARPD-based polymerorganic material, the water absorption of the material was studied as a function of the increase in the composition of polyethylene of the grade HPPE (granules) from 10 to 65% (Figure-3).

**Figure-2.** Dependence of water absorption of polymer organic material on the basis of ARPD on the amount of polyethylene.

The dependence of water absorption on the amount of polyethylene in the investigated range from 10 to 60% was practically linear, reaching a minimum at the point corresponding to the water absorption of a sample containing 60% polyethylene. The increase in the amount of polyethylene to 65% in the investigated compositions

based on the ARPD resulted in a sharp increase in water absorption, which is explained by the insufficient amount of binder (ARPD) and, as a result, the increased content of pores. The water absorption in the composition with the minimum amount of polyethylene is 0.1% and is doubled with 60% polyethylene content.



The results of the studies showed that the strength properties of the polymerorganic material based on ARPD depend on the mass content of the polymer material, while the water resistance value does not change significantly. Therefore, the main index determining the optimal composition of the material is the compressive strength.

Thus, the optimum composition of polymerorganic material with the maximum strength and minimum water absorption is the composition at the following mass content of components: ARPD - 40-50%, polyethylene waste - 50-60%.

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