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IMPROVEMENT OF OPERATIONAL AND REPAIR CHARACTERISTICS OF CARGO TANK SURFACES OF BULK TANKER-CHEMICAL CARGO VEHICLES BY IMPLEMENTING HYBRID EPOXY-POLYURETHANE COATINGS

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Summary

Introduction. *The tanker fleet plays a key role in the resource supply of the world's leading countries. At the same time, a significant part of the tanker surfaces is in contact with an aggressive environment, which leads to the course of corrosion processes and, as a result, a decrease in the strength of metal structures. This, in turn, can lead to tanker accidents and the risk of environmental pollution. Therefore, new hybrid paint coatings are needed for the reliable operation of the surfaces of ship cargo tanks.* ***Purpose.*** *Thus, the work aims to analyze the structure of polymer materials to create polymer coatings with optimal composition intended for protecting the surfaces of cargo tanks of bulk chemical tankers.* ***Results.*** *To create polymer coatings designed for the protection of ship cargo tanks, the ED-20 epoxy oligomer was selected as a binder, which was cured with hardeners based on aliphatic amines, in particular: polyethylene polyamine (PEPA), triethylenetetramine (TETA), canine 2609. The optimal content of ingredients for creating a hybrid epoxy coating was determined based on comprehensive studies of the structure and properties. To create a hybrid coating with optimal functional properties, it is advisable to use as a binder – epoxy oligomer ED-20 (100 pts.wt.), hardener polyethylene polyamine (10 pts.wt.), polyurethane varnish UR 294 (5-10 pts. wt.).* ***Conclusions.*** *Studying the structure and properties of the created materials made it possible to rationally combine the ingredients to obtain a hybrid coating with optimal properties. Based on the analysis of the obtained X-ray diffraction patterns of the studied samples, it was found that when polyurethane varnish UR 294 is introduced*

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in an amount of (5-10 pts.wt.) into the composition of the epoxy polymer, the average Bragg distance between the layers of molecular links of the epoxy polymer and PEPA increases. At the same time, the impact strength increases from 7.0 kJ/m² (epoxy matrix) to 11.1-12.0 kJ/m² (hybrid epoxy-polyurethane matrix). The developed hybrid polymer coatings can be used to improve the operational characteristics of cargo tanks of bulk chemical tankers.

Key words: *ship cargo tanks, epoxy oligomer, polyurethane varnish, hybrid coating, structure, impact strength.*

ПІДВИЩЕННЯ ЕКСПЛУАТАЦІЙНО-РЕМОНТНИХ ХАРАКТЕРИСТИК ПОВЕРХОНЬ ВАНТАЖНИХ ТАНКІВ НАЛИВНИХ ТАНКЕРІВ-ХІМОВОЗІВ ШЛЯХОМ ВПРОВАДЖЕННЯ ГІБРИДНИХ ЕПОКСИ-ПОЛІУРЕТАНОВИХ ПОКРИТТІВ

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Анотація

Вступ. *Танкерний флот відіграє ключову роль у ресурсозабезпеченні провідних країн світу. При цьому значна частина поверхонь танкерів контактує з агресивним середовищем, що призводить до корозійних процесів та, як наслідок, зниження міцності металоконструкцій. Це своєю чергою може призвести до аварії танків та ризику забруднення навколишнього середовища. Тому для надійної експлуатації поверхонь суднових вантажних танкерів виникає необхідність створення нових, гібридних лакофарбових покриттів. Отже,* ***метою*** *роботи є проведення аналізу структури полімерних матеріалів для створення оптимальних за складом полімерних покриттів, призначених для захисту поверхонь вантажних танків наливних танкерів-хімовозів.* ***Результати.*** *Для створення полімерних покриттів, призначених для захисту суднових вантажних танків, у якості зв'язувача обрано епоксидний олігомер ЕД-20, який затверджували твердниками на основі аліфатичних амінів, зокрема: поліетиленполіамін (ПЕПА), триетилентетрамін (ТЕТА), Ancatine 2609. На основі комплексних досліджень структури і властивостей визначено оптимальний вміст інгредієнтів для створення гібридного епоксидного покриття. Для створення оптимального за функціональними властивостями* *гібридного покриття доцільно використовувати у якості зв'язувача епоксидний олігомер ЕД-20 (100 мас.ч.), твердник поліетиленполіамін (10 мас.ч.), поліуретановий лак УР 294 (5–10 мас.ч.).* ***Висновки.*** *Дослідження структури і властивостей створених матеріалів дало змогу раціонально поєднати інгредієнти для отримання гібридного покриття з оптимальними властивостями. На основі аналізу отриманих рентгенівських дифрактограм досліджуваних зразків виявлено, що при введені поліуретанового лаку УР 294 у кількості (5–10 мас.ч.) до складу епоксидного полімеру відбувається зростання середньої Бреггівської відстані між шарами молекулярних ланок епоксидного полімеру та ПЕПА. При цьому відбувається підвищення ударної в'язкості з 7,0 кДж/м^2 (епоксидна матриця) до 11,1–12,0 кДж/м^2 (гібридна епокси-поліуретанова матриця). Розроблені гібридні полімерні покриття можливо використовувати для підвищення експлуатаційних характеристик вантажних танків наливних танкерів-хімовозів.*

Ключові слова: *суднові вантажні танки, епоксидний олігомер, поліуретановий лак, гібридне покриття, структура, ударна в'язкість.*

Introduction. Tankers designed to transport oil and petroleum products play a strategic role and affect the resource supply of many countries. At the same time, bulk chemical tankers transporting oil and petroleum products face the problem of corrosion destruction of cargo tanks. Corrosion destruction leads not only to a decrease in the strength of structural materials but can also cause tanker accidents, lead to economic losses, and pose a risk to environmental pollution and vessel safety [1].

Statement of the problem. During the operation of cargo tanks of bulk chemical tankers, three main types of corrosion destruction are distinguished: general corrosion (the presence of a uniformly distributed layer of rust on the metal surface, which is a consequence of atmospheric exposure), pitting corrosion (the presence of deep defects in the form of cavities or pits that develop on the horizontal surfaces of cargo tanks), pitting corrosion (the presence of deep point damage, pitting) [2-4]. When transporting petroleum products, the lower part of the tanks is in contact with the cargo. The most aggressive component in crude oils is hydrogen sulfide (H_2S). Hydrogen sulfide is corrosive to steel, so pitting corrosion is possible. It should also be noted that high levels of free sulfur are formed near the breathing valves of cargo tanks, where there is a greater chance of oxygen entering. Sulfur, combined with water and oxygen, can lead to the formation of sulfuric acid (similar to the formation of sulfur oxide in an inert gas), which in turn leads to pitting corrosion in the lower part of the tank. Since the upper part of the tank is in contact with the gaseous atmosphere, general corrosion is possible [5, 6].

Analysis of recent research and publications. The protection of the surfaces of cargo tanks of bulk tankers from corrosion is generally a set of measures aimed at preventing and inhibiting corrosion processes, preserving and maintaining their performance during their operation. A combined method is the most effective method of protecting metal structures from corrosion. According to the requirements of the IMO (May 2010) [7], regarding the protection against corrosion of cargo oil tanks of tankers for the transportation of crude oil, the combined method involves the use of alloying elements for steels and the use of protective coatings, respectively. At the same time, using protective coatings on a polymer basis is relevant, and the list of these is quite significant. Their use is due to high adhesive [8, 9], physical and mechanical [10, 11] properties, and resistance to the influence of aggressive external factors [12-14]. Therefore, when creating new polymer coatings intended to protect the surfaces of bulk chemical tank cargo tankers, selecting ingredients and considering their structural features and operating conditions is advisable.

Purpose: Analyzing the structure of polymer materials to create polymer coatings with an optimal composition intended to protect the surfaces of cargo tanks of bulk chemical tankers.

Presentation of the main material.

Materials and methods. To create polymer coatings intended for the protection of the surfaces of cargo tanks of bulk chemical tankers, the following ingredients were selected: binder-epoxy oligomer ED-20 (ISO 18280:2010), which was cured with various hardeners based on aliphatic amines, in particular, polyethylene polyamine (PEPA), triethylenetetramine (TETA), canine 2609. Among the most common hardeners (aromatic amines, acids, and anhydrides), aliphatic amines are characterized by an increased polymerization reaction rate at room temperature, determining their choice for experimental studies. Using aromatic amine hardeners, acids, and anhydrides involves elevated temperatures and increased time needed for the thermal crosslinking process.

Polyethylenepolyamine PEPA (TU 6-05-241-202-78) is an amine-type hardener that can cure materials at room temperature.

Triethylenetetramine TETA (TU 6-02-1099-83) is a cold-curing hardener that differs slightly from PEPA in the presence of amide groups.

Ancamine 2609 (Zhuzhou Polymer Co., Ltd., China). The hardener does not contain nonyl phenol. It is characterized by low viscosity, moisture resistance, and increased reactivity at low temperatures (268 K).

Polyurethane varnish UR 294 (TU 6-10-1462-84) was used to improve the properties of epoxy coatings. UR 294 results from the reaction of isocyanates (R–N=C=O) with reactive water molecules or unstable hydrogen atoms. It is characterized by resistance to the effects of dilute alkali solutions, weak acids, solvents, fuels and lubricants, and gasoline.

The formation of polymer materials was carried out according to works [11-13], with the following ratio of ingredients:

1) epoxy oligomer ED-20 (100 pts.wt.) + hardener polyethylenepolyamine (10 pts.wt.);
2) epoxy oligomer ED-20 (100 pts.wt.) + hardener triethylenetetramine (10 pts.wt.);
3) epoxy oligomer ED-20 (100 pts.wt.) + hardener Ancamine 2609 (10 pts.wt.).

The structural organization of polymers cured with different hardeners was studied by wide-angle X-ray diffraction on an XRD-7000 diffractometer (Shimadzu, Japan), the X-ray optical scheme of which was performed according to the Debye–Scherrer method for the passage of the primary beam through the sample under study, using CuK_{α}-radiation (λ = 1.54 Å) and a graphite monochromator [14]. The analyses were performed by automatic step-by-step scanning in the U = 30 kW, I = 30 mA mode in the scattering angle range (2θ) from 3.0 to 40 degrees, the exposure time was 5 s. The temperature of the studies was T = 293±2 K.

The surface images of the samples were taken on a scanning electron microscope REMM-102 AT SELMI manufactured in Sumy (Ukraine) (magnification ×800 times, operating voltage 15 kW) in the secondary electron mode. To obtain more precise information about the morphology of the fracture surface, the test samples were coated with carbon, and the coating thickness was 10–20 nm before the study.

Analysis of properties and structure of polymer materials. Based on the analysis of wide-angle X-ray diffraction patterns of the starting materials, it was found that the studied epoxy matrices polymerized with different hardeners have an amorphous structure (Fig. 1). According to the ratio of ED-20 + PEPA used when obtaining a network polymer, the central diffraction maximum ($2\theta_{max} \approx 18.1$) indicates the existence of a close order during the translation in space of fragments of interstitial molecular links of ED-20, and the secondary maximum ($2\theta_{max} \approx 5.4$o) is characteristic of the PEPA hardener. The average value of the period d of close ordering of both types of interstitial molecular links (the average distance between the layers of these links in the volume) was determined according to the Wolf–Bragg equation:

$$d = \lambda(2\sin\theta m)-1,$$

де λ – the wavelength of characteristic X-rays (λ = 1.54 Å for CuKα– rays), are 4.89 Å and 16.34 Å, respectively.

The use of other hardeners (TETA and Ancamine 2609) during thermal crosslinking of the epoxy binder ED-20 provides a shift of the central diffraction maximum in intensity, which characterizes the features of the ED-20 structure to the region of smaller angles from $2\theta_{max} \approx 18.1$o (for the hardener PEPA) to $2\theta_{max} \approx 17.8$o (for the hardener TETA) and $2\theta_{max} \approx 17.6$o (for the hardener Ancamine 2609). This indicates that as a result of the formation of epoxy polymers using different hardeners, the average Bragg distance between the layers of molecular links of the epoxy matrices is:

– *d* ≈ 4.89 Å – for PEPA hardener;
– *d* ≈ 4.97 Å – for TETA hardener;
– *d* ≈ 5.03 Å – for Ancamine 2609 hardener.

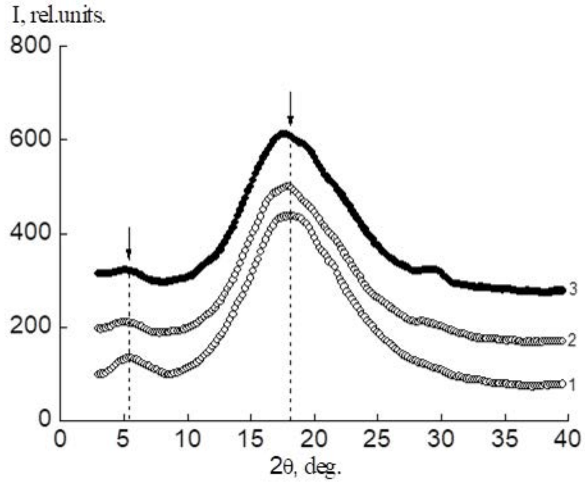
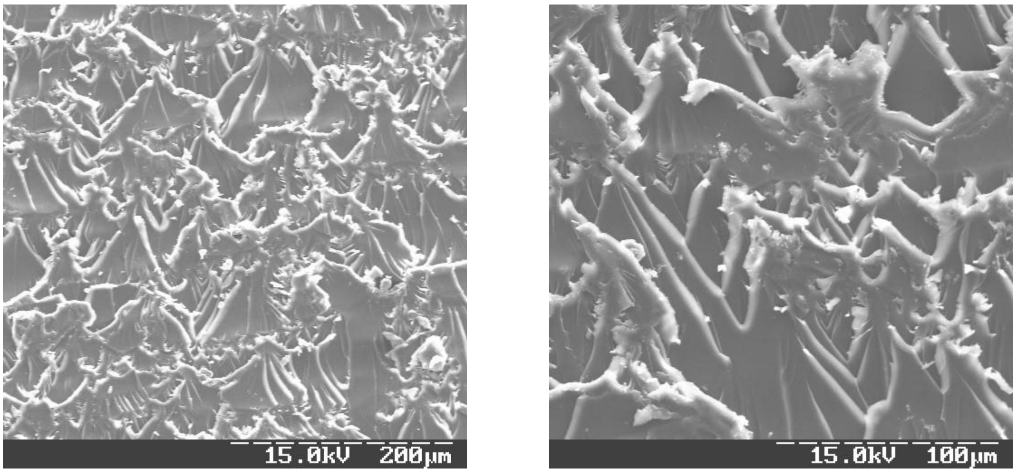


Fig. 1. Wide-angle X-ray diffraction patterns of epoxy samples cured with PEPA (1), TETA (2), and Ancamine 2609 (3)

It should be noted that when using different hardeners, the low-intensity diffraction maximum, which characterizes the features of their structure, has a different angular position. Accordingly, the average distance between the layers of PEPA units in the polymer volume is 16.34 Å, for TETA and Ancamine 2609 this value is 17.24 Å, respectively.

Thus, from the obtained results, it is possible to conclude that the smaller the average Bragg distance between the macromolecular chains of the epoxy resin during its curing, the greater the packing density of the macromolecular chains of the polymer. Therefore, a polymer matrix based on the epoxy oligomer ED-20 (100 pts.wt.) and the hardener PEPA (10 pts.wt.) was chosen for further experimental studies.

Analysis of the fracture surface of the polymer matrix at different magnifications (Fig. 2, a, b) allowed us to identify the mesh structure of the matrix, characterized by the uniformity of the facets. This indicates a uniform distribution of internal stresses and, consequently, the stability of the operational characteristics of such materials.



a) b)

Fig. 2. Morphology of the epoxy matrix based on the epoxy oligomer ED-20 (100 pts.wt.) and the hardener PEPA (10 pts.wt.)

It should be noted that polymer coatings based on epoxy binders are characterized by improved adhesive and physicomechanical and stable chemical properties, which allows them to be used to protect vehicles, including the surfaces of cargo tanks of bulk chemical tankers. However, such coatings may contain structural defects in the form of pores. Such defects lead to the absorption and migration of water molecules and other substances (including oil product molecules) at the “coating-substrate” interface, which leads to corrosion of metal surfaces and delamination of coatings. In this case, the barrier characteristics of epoxy coatings can be improved by including a second phase, a liquid phase (in the form of a polyurethane binder), which reduces or inhibits the diffusion process of oxygen and molecules in an aggressive environment. Such an effect is associated with forming physical interpenetration of dissimilar binders without forming covalent bonds between polymer networks. At the same time, the polyurethane coating is characterized by low resistance to aggressive environments, particularly oil products.

The development of hybrid coatings based on epoxy and polyurethane binders is relevant to ensure barrier protection of the surfaces of cargo tanks of bulk chemical tankers. Therefore, at the next stage, the features of the change in the structure of polymers based on the epoxy binder ED-20 and the hardener PEPA, into the volume of which polyurethane varnish UR 294 was introduced in different mass ratios, were studied.

Based on the analysis of wide-angle X-ray diffraction patterns of the studied samples (Fig. 3), it was found that when q = 5 pts.wt. and q = 10 pts.wt. of polyurethane varnish UR 294 were added to the volume of epoxy polymer, a shift was observed in both the main and secondary diffraction maxima in intensity, which characterize the structure of epoxy resin ED-20 and hardener PEPA, to the region of smaller angles (Fig. 3, curves 1-3). At the same time, the average Breg distance between macromolecular chains tends to increase for ED-20 from 4.89 Å (0 pts.wt.) to 4.92 Å (5 pts.wt.) and 5.00 Å (10 pts.wt.), and increases significantly for the PEPA hardener from 16.34 Å (0 pts.wt.) to 17.7 Å (5 pts.wt.) and 18.78 Å (10 pts.wt.).

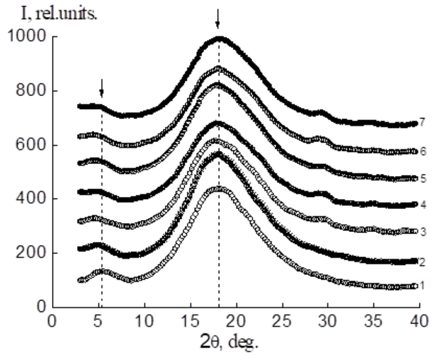


Fig. 3. Wide-angle X-ray diffraction patterns of epoxy polymer (1) modified with polyurethane varnish UR 294: 5 pts.wt. (2), 10 pts.wt. (3), 15 pts.wt. (4), 20 pts.wt. (5), 25 pts.wt. (6), 30 pts.wt. (7)

Further, an increase in the content of polyurethane varnish (q = 15-30 pts.wt.) in the epoxy polymer composition leads to a decrease in the average Bragg distance between the layers of ED-20 and PEPA molecular units compared to the sample containing 10 wt. parts of filler (Fig. 3, curves 4-7, table 1).

Table 1

Breg distance between molecular units of samples

The content of UR 294 in the volume of the epoxy matrix, q, pts.wt.	$2\theta_m$, deg.	d, Å (ED-20)	$2\theta_m$, deg. (ED-20)	d, Å (PEPA)
0	18.1	4.89	5.4	16.34
5	18.0	4.92	5.0	17.70
10	17.7	5.00	4.7	18.78
15	17.9	4.95	5.1	17.32
20	17.9	4.95	5.1	17.32
25	18.1	4.89	5.1	17.32
30	18.2	4.86	5.1	17.32

Thus, by changing the content of polyurethane varnish UR 294 in the volume of the epoxy matrix, we observed a change in the structure and, therefore, the physical and mechanical properties of hybrid epoxy-polyurethane coatings (Fig. 4).

It is proven that the introduction of polyurethane varnishes UR 294 at a content of q = 5-10 pts.wt. provides a monotonic increase in impact strength from W = 7 kJ/m^2 to W = 11.1-12.0 kJ/m^2. It was assumed that polyurethane varnish UR-294, which has isocyanate groups (R–N=C=O), can interact with the epoxy oligomer's hydroxyl groups, which increases impact strength by 1.5-1.7 times. The increase in mechanical characteristics is primarily associated with chemical interaction, which affects structural changes – a change in the length between the layers of molecular links consistent with the results of wide-angle radiography (Fig. 3).

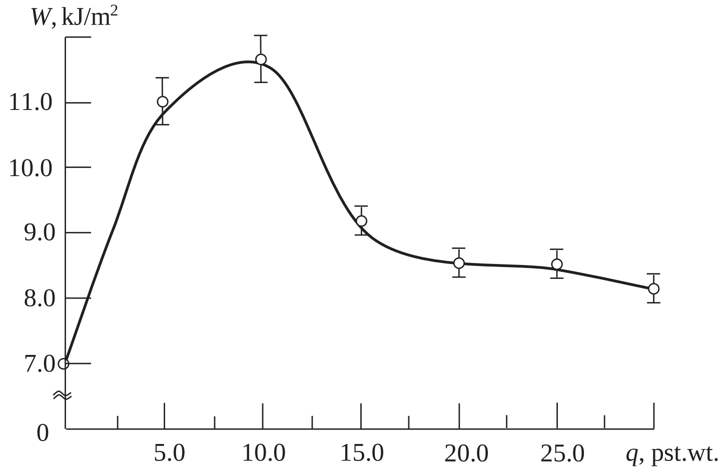
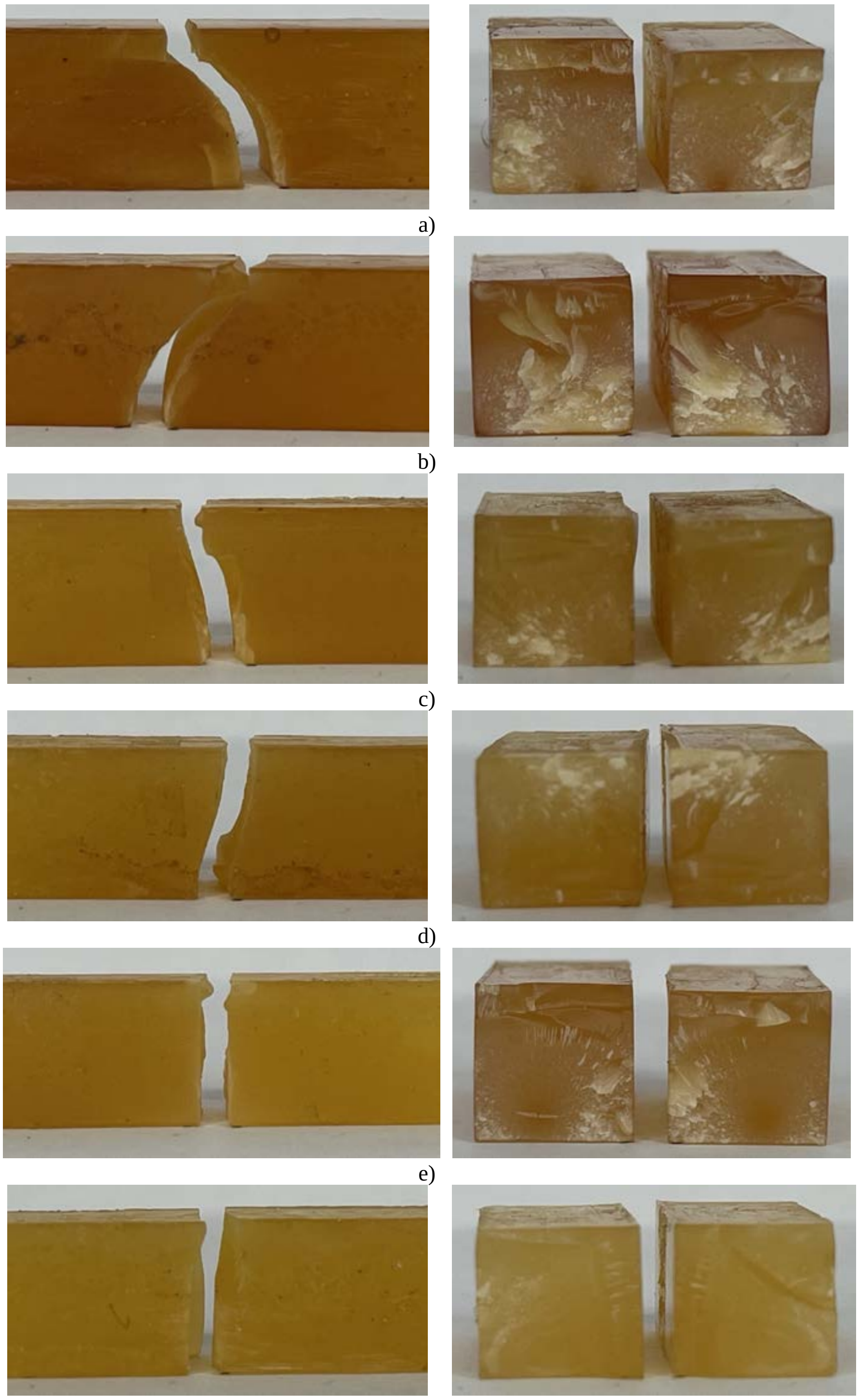


Fig. 4. Dependence of the impact strength (W) of the hybrid polymer matrix on the content of polyurethane varnish UR-294

The following was observed when forming experimental samples with different polyurethane varnish contents. Unlike epoxy samples with a content of q = 5-15 pts.wt. of UR 294, which were polymerized at room temperature T = 293 K for τ = 12 h, samples with a content of q = 20-30 pts.wt. of UR 294 were partially polymerized (under the same conditions). The obtained effect can be explained by the fact that polyurethane varnish UR 294 at an increased content prevents the hardening of the epoxy binder. This is due to the presence of flexible polymer macromolecules and chain segments in the molecular structure and the structure of the branched chain. Thus, the contact area between the epoxy ring and the hardener is reduced, so the complete hardening of the polymer occurred after thermal treatment under previously established temperature-time conditions [11-13].

Additionally, the nature of the microfracture of the surface of hybrid epoxy-polyurethane materials was analyzed (Fig. 5). The fracture surface of samples characterized by maximum impact toughness values was previously investigated (Fig. 5, a, b). A curved fracture trajectory of hybrid epoxy-polyurethane materials was established, which indicates an increase in the time and resistance to fracture under the influence of impact loading [15]. It was assumed that under the influence of an external load, the main epoxy chain deforms under



a)

b)

c)

d)

e)

f)

Fig. 5. Type of epoxy composites microfracture with different polyurethane varnish contents UR-294, q, wt. parts: a) 5.0; b) 10.0; c) 15.0; d) 20.0; e) 25.0; f) 30.0

the action of the applied force. Then, the interpenetrating chain of the polyurethane varnish provides a partial plasticizing effect. The synergistic effect in the form of rigidity (epoxy chain) and moderate plasticizing action (polyurethane varnish chain), when reaching maximum stresses in the polymer, provides rapid formation and growth of internal defects until the moment of fracture. In this case, a fracture surface was observed with areas of loose origin with a complex heterogeneous structure (Fig. 5, a, b).

Meanwhile, hybrid epoxy-polyurethane materials with a higher content of polyurethane varnish UR-294 (Fig. 5, c-f) showed a more gentle fracture. This is due to the ratio of epoxy binder and polyurethane varnish. In this case, the increase in the plasticizing effect of the polymer is directly related to the yield point, the decrease of which provides elastic-plastic deformation. At the same time, the fracture surface of such materials is characterized by structural heterogeneity and leads to a rapid, viscoelastic fracture in the region of maximum stresses. This affects the strength characteristics of hybrid materials.

Conclusion. Based on studies of the structure of epoxy matrices polymerized with various amine hardeners, it was found that to improve the operational and repair characteristics of the surfaces of cargo tanks of bulk chemical tankers, it is advisable to use a polymer matrix based on the epoxy oligomer ED-20 (100 pts.wt.) and the hardener PEPA (10 pts.wt.). Analysis of the morphology of the polymer matrix allowed us to identify its homogeneity. In such materials, the smallest (among the studied matrices) average Bragg distance between the layers of molecular links of the matrix was observed – $d \approx 4.89$ Å. This indicates a densification of the polymer network, which provides improved material characteristics.

The mechanical strength and fracture pattern of epoxy matrices modified with polyurethane varnish UR-294 under impact loading were studied. It was found that the maximum impact strength value – $W = 12.0$ kJ/m^2 is characteristic of hybrid coatings containing epoxy oligomer ED-20 – 100 pts.wt., hardener polyethylenepolyamine – 10 pts.wt., polyurethane varnish UR 294 – 10 pts.wt. Microfracture analysis of hybrid epoxy-polyurethane materials obtained using optical microscopy allows us to state that the increase in impact strength is associated with the chemical interaction of isocyanate groups of polyurethane varnish with hydroxyl groups of the epoxy oligomer. Due to the presence of flexible macromolecules and branched polymer segments of polyurethane varnish UR 294 in the material structure, the contact area between the epoxy ring and the hardener decreases, slowing the hardening process. A plasticizing effect of polyurethane varnish is created, affecting cracks' growth and propagation. In this case, the hybrid polymer's relatively high fracture energy is required for crack propagation. This makes it possible to use the developed hybrid epoxy-polyurethane coatings to protect the surfaces of cargo tanks of bulk chemical tankers.

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