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Development of a polymer matrix with improved performance characteristics for protection of vehicle elements

[1]M. V. Brailo, [1]A. V. Buketov, [1]S. V. Yakushchenko, [1]A. V. Sapronova, [2]O. S. Kobelnyk, [3]I. T. Yarema, [4]I. P. Fesenko

[1] Kherson State Maritime Academy; 20, Ushakova Avenue, Kherson, 73000, Ukraine

[2] Technical College of Ternopil National Technical University named after Ivan Pulyuy; General Tarnovsky Str., Ternopil, 746024, Ukraine

[3] Ternopil National Technical University named after Ivan Pulyuy; 56, Ruska Str., Ternopil, 46000, Ukraine

[4] V. Bakul Institute for Superhard Materials of National Academy of Ukraine; 2, Avtozavodska Str., Kyiv, 04074, Ukraine

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Abstract

The impact of Norsodyne O 12335 AL polyester resin content in epoxy oligomer ED-20 in terms of thermophysical properties has been investigated. The dynamics of the heat resistance index (according to Martens) of the composite with the increasing content of polyester resin in the epoxy oligomer to q = 120 weight parts and the optimal polyester binder content have been established. The change of thermal coefficient of linear expansion of epoxy-polyester composite in different temperature ranges and linear shrinkage from polyester resin content have been studied. It has been experimentally proved that with the introduction of Norsodyne O 12335 AL in the amount of q = 10–20 weight parts a composite material is formed, which is characterized by the minimum values of thermal coefficient of linear expansion in the following ranges: in the range of ΔT = 303–323 K – $\alpha = 1.6 \cdot 10^{-5}$ K^{-1}, in the range of ΔT = 303–373 K – $\alpha = (2.0–2.5)\cdot 10^{-5}$ K^{-1}, in the range of ΔT = 303–423 K – $\alpha = (3.8–3.9)\cdot 10^{-5}$ K^{-1}, in the range of ΔT = 303–473 K – $\alpha = (8.8–8.9)\cdot 10^{-5}$ K^{-1}. At the same time indicators of linear shrinkage decrease in comparison with an epoxy matrix from Δl = 0.32 % to Δl = 0.13–0.14 %. It has been analyzed that the obtained values of thermal coefficient of linear expansion and linear shrinkage are correlated with the indicators of heat resistance (according to Martens) and with the previously studied values of physical and mechanical properties. The composition of the epoxy-polyester matrix has been established, which in the complex differs in the increased indicators of thermophysical properties. Based on the obtained results, an epoxy-polyester matrix has been developed, which is proposed to be used in the formation of protective coatings for parts of vehicles operating under the influence of variable and elevated temperatures.

Keywords: *composite material, epoxy-polyester matrix, heat resistance (according to Martens), means of transport, protective coating, shrinkage, thermal coefficient of linear expansion, thermophysical properties.*

Introduction

Nowadays, oil and gas, which are important energy resources, are transported in several ways. One of the main ones is the use of land transport, in particular pipeline and water transport (oil tankers and gas carriers). The technical condition of vehicles and their operational characteristics significantly affect the economic efficiency of transportation. Improving the performance and ensuring a reliable technical condition is due to the use of structural materials, which are made of metals and alloys of different quality in the course of vehicles parts manufacturing elements [1–4]. These elements, both in pipelines and on vessels, are exposed to aggressive environments, which leads to corrosion and failure. Polymeric protective coatings are used to prevent corrosion on the metal and reduce its interaction with aggressive environments [2–7]. There are many different coatings for different functional purposes, which differ in composition and cost [2–3, 8–13]. At the same time, it is important when creating polymer coatings to take into account the influence of deformations and temperature changes, which have a negative effect on both the metal and the coating as a whole. Therefore, it is important to create a protective coating based on a polymer matrix, which will be characterised not only by improved physical and mechanical properties, but also by thermophysical properties.

With the development of modern technologies, the requirements for protective materials are increasing. It is

* Corresponding author: mv.brailo@gmail.com

known that polymeric protective coatings having an epoxy base have become widely used [2, 5–7, 11–14]. To improve the properties of epoxy-based protective coatings, modifiers, plasticizers, nano-, micro- and disperse fillers of different nature are introduced into their composition, as well as treatment with different energy fields [8–12, 16–19]. At the same time, polymeric materials based on polyester resins are widely used [8, 10, 14–15]. In previous works [16–17] an epoxy-polyester matrix was developed, which in the complex differs in improved indicators of physical and mechanical properties. Therefore, the creation of an epoxy-polyester matrix to protect the elements of vehicles, which will have improved performance under the influence of variable and elevated temperatures is a challenging issue.

The purpose of the work is to develop a polymer matrix with improved performance characteristics to protect the elements of vehicles.

Research materials and methods

In order to form a matrix for CM with improved thermophysical properties, the following ingredients were used.

A low molecular weight epoxy diane oligomer of the ED-20 grade (q = 100 weight parts) was chosen as the main component for the binder. It should be noted that the molecules of epoxy oligomers contain glycidyl and epoxy groups, which are able to interact with the hardener to form a crosslinked structure in the materials in the form of a grid [2].

Ortho-phthalic unsaturated pre-accelerated polyester resin (PR) Norsodyne O 12335 AL.

Hardener of cold hardening of epoxy resins polyethylene polyamine (PEPA) (TU 6-05-241-202–78).

Hardener for polyester resins – Butanox-M50, which is a methyl ethyl ketone peroxide.

In order to determine the optimal ratio between the concentration of epoxy and polyester resins in the binder, the physical and mechanical properties of CM were studied. The ratio of the concentration of polyester resin Norsodyne O 12335 AL varied within: q = 0–120 weight parts per 100 weight parts of epoxy oligomer ED-20. The concentration of hardeners in the compositions and the crosslinking temperature were determined in accordance with previous research results.

The following thermophysical properties were studied: heat resistance (according to Martens), thermal coefficient of linear expansion.

Heat resistance (according to Martens) of CM was determined according to GOST 21341–75. The method of research is to determine the temperature at which the test specimen was heated at a rate of υ = 3 K/min under the impact of a constant bending load F = 5.0 ± 0.5 MPa, as a result of which it is deformed to a given value (h = 6 mm).

The thermal coefficient of linear expansion of the specimens was calculated from the curve of relative deformation to temperature, approximating this dependence by an exponential function. The relative deformation was determined by the change in the length of the specimen with increasing temperature in stationary conditions (GOST 15173–70). The dimensions of the specimens for the study: 65×12×12 mm, the non-parallelism of the ground ends was not more than 0.02 mm. Before the study, the length of the specimens was measured with an accuracy of ± 0.01 mm. The heating rate was υ = 2 K/min.

Materials were hardened according to the experimentally established mode: the formation of specimens and their aging over time t = 12.0 ± 0.1 h at a temperature T = 293 ± 2 K, heating at a rate of υ = 3 K/min to a temperature T = 393 ± 2 K, keeping the specimens at a given temperature for a time t = 2.00 ± 0.05 h, slow cooling to a temperature T = 293 ± 2 K. In order to stabilize the structural processes in the matrix, the specimens were kept for a time t = 24 h in air at a temperature T = 293 ± 2 K with the following experimental tests.

Research results and their discussion

According to preliminary research results [20], it was found that to form a composite material with improved physical and mechanical properties, it is necessary to introduce components in the following ratio: epoxy diane oligomer ED-20 (q = 100 weight parts) + polyester resin Norsodyne O 12335 AL (q = 20 weight parts per 100 weight parts of epoxy oligomer ED-20) + PEPA hardener (q = 10 weight parts per 100 weight parts of ED-20) + hardener for polyester resins – Butanox-M50 (q = 1 weight part per 100 weight parts of Norsodyne O 12335 AL).

It is important to investigate the effect of the content of polyester resin Norsodyne O 12335 AL in the epoxy oligomer ED-20 on the thermophysical properties of CM.

At the initial stage, the heat resistance (according to Martens) of CM with different content of polyester binder was investigated (Fig. 1). It was found that when adding polyester resin Norsodyne O 12335 AL in epoxy oligomer ED-20 heat resistance (according to Martens) of the composite changes. With a content of q = 10 weight parts the values increase, but not significantly, from T = 338 K (for epoxy matrix) to T = 339 K. Subsequent introduction of a polyester binder leads to a decrease in the heat resistance of the material. The figures are gradually declining. With the content of Norsodyne O 12335 AL in the amount of q = 10 weight parts the value of heat resistance (according to Martens) is reduced to T = 335 K. Next, the values are reduced in direct proportion to the addition of polyester binder to T = 310–312 K at a content of q = 100–120 weight parts. According to the passport characteristics of polyester resin Norsodyne O 12335 AL it is known that the heat resistance of the approved material is T = 323 K. It was investigated that the heat resistance of the epoxy matrix based on epoxy oligomer ED-20, as it was mentioned above, is T = 338 K. That is why it is evident that in the course of introduction of the polyester binder in the epoxy oligomer, heat resistance of the formed multicomponent composite is decreasing in proportion to the introduction of the component, which has low performance of this property. It can be assumed that with increasing PR content, the radical copolymerization of styrene with reactive double bonds of polyester affects the reaction of epoxy and hydroxyl groups with polyethylene polyamine hardener, which reduces the frequency of the crosslinking grid, thus it is reducing the number of loops of a three-dimensional mesh structure per unit of volume. In particular, it should be noted that with a small content of PR in the amount of q = 10–20 weight parts, indicators of thermophysical properties of CM do not change significantly and at q = 10 weight parts increase by ΔT = 1 K. It is obvious that for a given content of PR in the epoxy oligomer, the effect of the styrene copolymerization reaction does not significantly affect the interaction of epoxy and hydroxide groups with polyethylene polyamine. And if concentration is q = 10 weight parts, it leads to the activation of free bonds and increasing the degree of crosslinking of the CM. Similar dynamics can be observed in the preliminary study of the physical and mechanical properties of the composite with the addition of polyester resin Norsodyne O 12335 AL to the epoxy binder ED-20 .

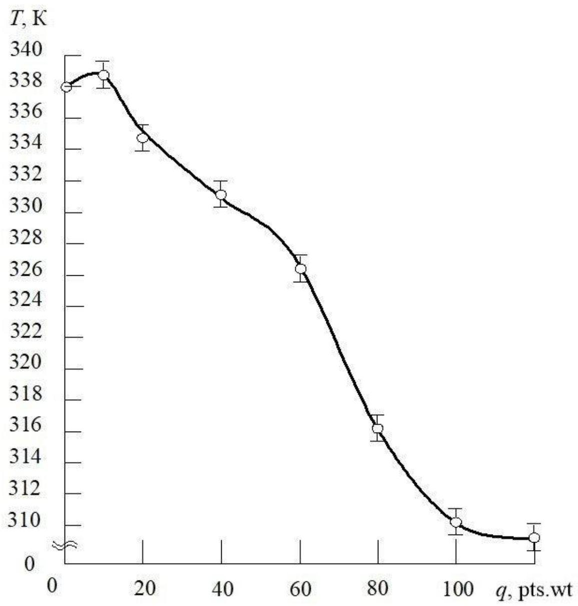
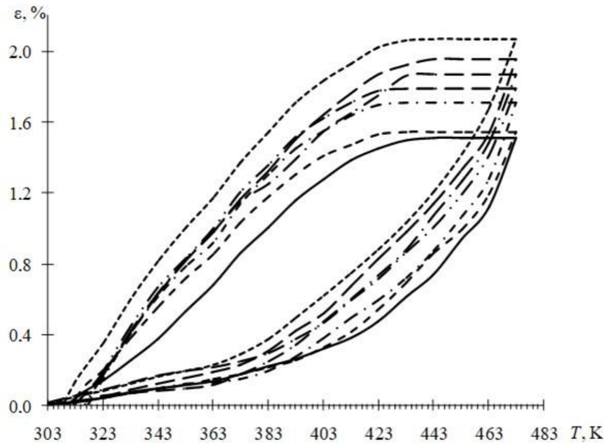


Figure 1 – Heat resistance (according to Martens) (*T*) of CM with different content of polyester resin (*q*) in the epoxy oligomer

At the next stage, on the basis of dilatometric curves (Fig. 2), the thermal coefficient of linear expansion (Table 1) of epoxy-polyester composites with different content of polyester resin in the binder and their linear shrinkage (Table 2) was investigated. The thermal coefficient of linear expansion of materials was analyzed in the following temperature ranges: 1 – ΔT = 303–323 K; 2 – ΔT = 303–373 K; 3 – ΔT = 303–423 K; 4 – ΔT = 303–473 K (Table 1). It has been defined that for the epoxy matrix the indicators of the thermal coefficient of linear expansion in different temperature ranges are as follows: $\alpha = 6.3 \cdot 10^{-5}$ K^{-1} (in the range of ΔT = 303–323 K), $\alpha = 6.8 \cdot 10^{-5}$ K^{-1} (in the range of ΔT = 303–373 K), $\alpha = 9.9 \cdot 10^{-5}$ K^{-1} (in the range of ΔT = 303–423 K), $\alpha = 10.9 \cdot 10^{-5}$ K^{-1} (in the range of ΔT = 303–473 K). In particular, according to the obtained data (Table 1), it is can be noticed that in the temperature ranges up to T = 423 K with the introduction of polyester resin into the epoxy oligomer, the indicators of thermal coefficient of linear expansion of materials are much lower if compared to the epoxy matrix.



——— – *(q) = (10,0) weight parts;*
- - - - – *(q) = (20,0) weight parts;*
— — — – *(q) = (40,0) weight parts;*
— · — · – *(q) = (60,0) weight parts;*
.... – *(q) = (80,0) weight parts;*
— · · — – *(q) = (100,0) weight parts;*
- · - · - – *(q) = (120,0) weight parts*

Figure 2 – Dilatometric curves of CM with different content of polyester resin in the epoxy oligomer

It is proved that at the content of Norsodyne O 12335 AL in the amount of q = 10– 20 weight parts, CM is formed, which is characterized by the minimum indicators of thermal coefficient of linear expansion in the following ranges: in the range of ΔT = 303–323 K – $\alpha = 1.6 \cdot 10^{-5}$ K^{-1}, in the range of ΔT = 303–373 K – $\alpha = (2.,0–2.5) \cdot 10^{-5}$ K^{-1}, in the range of ΔT = 303–423 K – $\alpha = (3.8–3.9) \cdot 10^{-5}$ K^{-1}, in the range of ΔT = 303–473 K – $\alpha = (8.8–8.9) \cdot 10^{-5}$ K^{-1}. Analyzing the linear shrinkage of materials at a given concentration of components (Table 2), it can be noticed that the composites also differ in the minimum shrinkage. The values decrease from Δl = 0.32 % (for epoxy matrix) to Δl = 0.13–0.14 % for the content of Norsodyne O 12335 AL in the amount of q = 10–20 weight parts. It is obvious that the abovementioned theory of interaction in the formation of epoxy and polyester binder in this composition leads to improved crosslinking of materials, which has the effect of increasing heat resistance (according to Martens), reducing thermal coefficient of linear expansion and linear shrinkage.

Increasing the content of PR to q = 40–100 weight parts leads to an increase in thermal coefficient of linear expansion in all studied temperature ranges (Table 1). At the same time in the range ΔT = 303–323 K values increase to $\alpha = (1.8–2.5) \cdot 10^{-5}$ K^{-1}, in the range of ΔT = 303–373 K – $\alpha = (3.3–4.1) \cdot 10^{-5}$ K^{-1}, in the range of ΔT = 303–423 K – $\alpha = (5.9–7.3) \cdot 10^{-5}$ K^{-1}, in the range of

Table 1 – Dependence of the thermal coefficient of linear expansion $\alpha \cdot 10^5$, K^{-1} on the content of polyester resin in different temperature ranges

The content of polyester resin *q*, weight parts	Temperature ranges *T*, K			
	303–323	303–373	303–423	303–473
0	6.3	6.8	9.9	10.9
10	1.6	2.5	3.9	8.9
20	1.6	2.0	3.8	8.8
40	1.8	3.3	5.9	10.5
60	2.0	3.4	6.1	10.9
80	2.5	4.1	7.3	11.1
100	2.4	3.5	5.9	10.5
120	2.0	2.1	4.9	10.1

Table 2 – Shrinkage rates of composites with different polyester resin content in epoxy oligomer

Characteristics	Epoxy matrix	Polyester resin *q*, weight parts						
		10	20	40	60	80	100	120
Shrinkage, *Δl*, %	0.32	0.13	0.14	0.27	0.30	0.31	0.35	0.39

ΔT = 303–473 K – α =(10.5–11.1)·10^{-5} K^{-1}. Also, in accordance with the results obtained in the study of linear shrinkage (Table 2), the dynamics of increasing its values to Δl = 0.27–0.35 % at the content of PR of q = 40–100 weight parts. In particular, the content of polyester resin in the amount of q = 120 weight parts, figures of thermal coefficient of linear expansion decrease to α =2.0·10^{-5} K^{-1} in the range of ΔT = 303–323 K, to α =2.1·10^{-5} K^{-1} in the range of ΔT = 303–373 K and up to α =4.9·10^{-5} K^{-1} in the range of ΔT = 303–423 K. The linear shrinkage is the maximum among the studied materials and is Δl = 0.27–0.35 %. It is known [40, 41, 165] that composites based on polyester resins are characterized by higher linear shrinkage compared to epoxy composites. It is obvious that at a high content of polyester binder when heating the material there are different temperature expansions of the crosslinked bonds of macromolecules of epoxy and polyester components, which leads to increased linear shrinkage of materials and reduced heat resistance (according to Martens). In particular, it can be assumed that there is a partial softening of materials at high temperatures, which leads to a decrease in thermal coefficient of linear expansion at a critical content. It should be noted that in the study of the physical and mechanical properties of CM with the introduction of Norsodyne O 12335 AL in the epoxy oligomer ED-20 over q = 40 weight parts deterioration of their indicators is proved. That is, the obtained values of thermal coefficient of linear expansion, linear shrinkage correlate with the indicators of heat resistance (according to Martens) and with the previously studied values of physical and mechanical properties.

Therefore, analyzing the results of the study of the thermophysical properties of CM with different polyester binder content in the epoxy oligomer, it can be stated that the introduction of an additional component generally leads to a decrease of heat resistance (according to Martens) of CM. In particular, it should be noted that at the content of q = 10 weight parts, heat resistance of the material increases, but not significantly. According to the obtained data of the thermal coefficient of linear expansion and linear shrinkage, it was found that the minimum values of the composites differ in the content of polyester binder in the amount of q = 10–20 weight parts, taking into account the results of experimental studies of the properties of CM, it can be argued that to create an epoxy-polyester matrix with improved thermophysical properties one should introduce polyester resin Norsodyne O 12335 AL (q = 10–20 weight parts) into the epoxy oligomer ED-20.

Conclusions

Based on the results of experimental studies, the following can be stated.

It has been found that the content of polyester resin Norsodyne O 12335 AL in the epoxy oligomer ED-20 affects the thermophysical properties of epoxy-polyester composite materials. It has been analyzed that at the content of q = 10 weight parts of polyester binder, the heat resistance (according to Martens) of the composite material increases, but not significantly, from T = 338 K (for epoxy matrix) to T = 339 K. It has been proved that the further content of Norsodyne O 12335 AL leads to a decrease in the heat resistance of the material. With the content of q = 100–120 weight parts of polyester, T = 310–311 K.

It has been experimentally investigated that the composite material differs by the minimum indicators of the thermal coefficient of linear expansion in all ranges and linear shrinkage if the content of Norsodyne O 12335 AL is q = 10–20 weight parts. Values of thermal coefficient of linear expansion are as follows: in the range of ΔT = 303–323 K – α =1.6·10^{-5} K^{-1}, in the range of ΔT = 303–373 K – α =(2.0–2.5)·10^{-5} K^{-1}, in the range of ΔT = 303–423 K – α =(3.8–3.9)·10^{-5} K^{-1}, in the range of ΔT = 303–473 K – α =(8.8–8.9)·10^{-5} K^{-1}. The values of linear shrinkage decrease from Δl = 0.32 % (for epoxy matrix) to Δl = 0.13–0.14 % if the content of Norsodyne O 12335 AL is q = 10–20 weight parts.

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УДК 667.64:678.026

Розробка полімерної матриці з поліпшеними експлуатаційними характеристиками для захисту елементів транспортних засобів

[1]М. В. Браїло, [1]А. В. Букетов, [1]С. В. Якущенко, [1]А. В. Сапронова, [2]О. С. Кобельник, [3]І. Т. Ярема, [4]І. П. Фесенко

[1] Херсонська державна морська академія;
20, проспект Ушакова, м. Херсон, 73000, Україна

[2] Технічний коледж Тернопільського національного технічного університету імені Івана Пулюя;
вул. Генерала Тарновського, м. Тернопіль, 746024, Україна

[3] Тернопільський національний технічний університет імені Івана Пулюя;
56, вул. Руська, м. Тернопіль, 46000, Україна

[4] Інститут надтвердих матеріалів ім. В.М. Бакуля НАН України;
2, вул. Автозаводська, м. Київ, 04074, Україна

У роботі досліджено вплив вмісту поліефірної смоли Norsodyne O 12335 AL у епоксидному олігомері ЕД-20 за показниками теплофізичних властивостей. Проаналізовано динаміку показника термостійкості (за Мартенсом) композиту при збільшенні вмісту поліефірної смоли у епоксидному олігомері до q = 120 мас.ч. та встановлено оптимальний вміст поліефірного зв’язувача. Досліджено зміну показників термічного коефіцієнту лінійного розширення (ТКЛР) епоксиполіефірного композиту у різних температурних діапазонах та лінійну усадку від вмісту поліефірної смоли. Експериментально доведено, що при введенні Norsodyne O 12335 AL у кількості q = 10–20 мас.ч. формується композитний матеріал, який характеризується мінімальними показниками ТКЛР у діапазонах: у області ΔT = 303–323 K – $\alpha = 1.6\cdot10^{-5}$ K^{-1}, у області ΔT = 303–373 K – $\alpha = (2.0–2.5)\cdot10^{-5}$ K^{-1}, у області ΔT = 303–423 K – $\alpha = (3.8–3.9)\cdot10^{-5}$ K^{-1}, у області ΔT = 303–473 K – $\alpha = (8.8–8.9)\cdot10^{-5}$ K^{-1}. При цьому показники лінійної усадки зменшуються порівняно з епоксидною матрицею від Δl = 0,32 % до Δl = 0,13–0,14 %. Проаналізовано, що отримані значення ТКЛР, лінійної усадки корелюють із показниками термостійкості (за Мартенсом) та із попередньо дослідженими значеннями фізико-механічних властивостей. Встановлено склад епоксиполіефірної матриці, який у комплексі відрізняється підвищеними показниками теплофізичних властивостей. На основі отриманих результатів розроблено епоксиполіефірну матрицю, яку запропоновано використовувати при формуванні захисних покриттів для елементів засобів транспорту, що експлуатуються в умовах впливу змінних та підвищених температур.

Ключові слова: *епоксиполіефірна матриця, засоби транспорту, захисне покриття, композитний матеріал, термостійкість (за Мартенсом), теплофізичні властивості, термічний коефіцієнт лінійного розширення, усадка.*