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OPTIMIZATION OF THE COMPOSITION OF ALUMINUM PHOSPHATE AND WALNUT SHELL-BASED COMPOSITION TO INCREASE THE CORROSION RESISTANCE OF PAINT COATINGS

The object of the study is anti-corrosion properties of walnut shell powder and aluminum phosphate mixtures. The existing problem is that the most effective chromate-containing pigments, which were traditionally used for the manufacture of paint and varnish coatings, are toxic. Given this fact, the research of scientists aims at finding alternative low-toxic compounds, which are phosphate pigments. Since they are inferior in efficiency, modern research is aimed either at the synthesis of new modifications and complex forms of pigments, or at the development of effective mixtures of pigments that would provide the necessary level of anti-corrosion protection of steel. Along with this, an urgent direction is to increase the level of environmental friendliness of paint and varnish coatings using annually renewable plant waste, which, due to the content of tannins, have proven themselves well for surface preparation before painting. The work investigated the effect of a mixture of non-toxic aluminum phosphate and finely ground walnut shell powder on the corrosion behavior of steel. An adequate mathematical model "composition – mass corrosion index" was proposed. The mathematical model allowed to establish the relationship between the composition and corrosion rate and find the optimal composition of the studied mixture. The calculations showed that at a ratio of aluminum phosphate and walnut shell powder of about 8:1, the mass corrosion rate of steel in the obtained extract is 0.020 g/($m^2 \cdot h$). By experimentally verifying the optimal composition of the study, a mass corrosion index of 0.018 g/($m^2 \cdot h$) was achieved, which confirms the theoretical calculations and ensures the practical applicability of the results obtained.

The results of the study will be useful for specialists working in the field of developing water-based anti-corrosion paints and coatings, with an emphasis on studying the influence of pigments and fillers on the corrosion behavior of steel.

Keywords: *aluminum phosphate, walnut shell, pigment, corrosion, mathematical model of corrosion rate.*

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1. Introduction

One of the important problems is the protection against corrosion of metal products and metal structures operated in the atmosphere and aggressive environments. This problem is solved by applying paint coatings that provide protection against corrosion for several years. The protective ability of paint coatings applied as the first layer on a metal surface, namely paint primers, is largely determined by a set of pigments and fillers, the extractive part of which, getting on the metal surface, provides inhibition of corrosion processes under the coating film.

One of the current directions in the development of modern paint and varnish materials is the creation of water-based coatings, which solves the issue of reducing the load on the environment by replacing oxic organic solvents with less aggressive and safe components.

The second direction in the development of paint and varnish coatings in recent decades is the complete replacement of traditionally used and highly effective chromate-containing pigments, which are extremely toxic, with more environmentally friendly compounds. The article [1] shows that there are no known industrial pigments that would be comparable in effectiveness to chromates. Therefore, the efforts of scientific groups in the world focused on finding effective mixtures of traditional pigments or synthesizing more effective compounds.

Chromates were replaced by numerous phosphate-based pigments, which were less aggressive and toxic pigments. Obviously, being inferior to toxic chromates, phosphate pigments needed to enhance their anti-corrosion ability. The solution to this issue lay in the selection of other fillers or pigments, which, when used together, would form synergistic mixtures with a higher level of anti-corrosion efficiency due to the mutual enhancement of the inhibitory effect. Another way is the synthesis of new pigments, their combinations and forms.

In the work [2], a number of combined and modified phosphate pigments containing Mn, Al, Zn, Cr and Mo were synthesized. The studies have established rather high anti-corrosion properties of the synthesized phosphates in comparison with traditional zinc phosphate. The best anti-corrosion properties were demonstrated by the combined pigment $AlPO_4/Al(PO_3)_3$, which was associated, according to the authors, with low solubility (0.54%) and the ability to passivate steel. The article [3] shows that increased anti-corrosion efficiency of phosphates, for example, aluminum phosphate, can be achieved by using it in the coating in a nano-sized state. According to the results of the studies, nano-sized aluminum phosphate surpassed the micronized known aluminum-zinc phosphate pigment. The effectiveness of nano-sized aluminum phosphate in the composition of a solvent-based polyurethane coating, commercial acrylic resin and a hardener such as isophorone diisocyanate has been confirmed by a number of accelerated anti-corrosion tests. In particular, the evaluation of adhesion, stability in the salt spray chamber, and impedancemetry of painted samples with a coating thickness of up to 220 μm when exposed to a sodium chloride solution was carried out.

Another interesting approach was the synthesis of environmentally friendly highly effective anti-corrosion pigments obtained by the core-shell principle. The authors of the work [4] synthesized a pigment based on a less expensive filler pigment (aluminum oxide) as a core and a shell based on magnesium and zinc phosphates, which is superior to individual phosphate compounds. The effectiveness of corrosion inhibition of paint and varnish compositions increased with increasing concentration of core-shell pigments with maximum efficiency at a concentration of 15%.

The combination of phosphate pigments into synergistic mixtures is promising. Thus, in the work [5], a synergistic mixture of zinc orthophosphate hydrate, aluminum, molybdenum and zinc orthophosphate silicate, calcium, strontium, aluminum was studied in comparison with zinc phosphate, which has recently been considered dangerous for the environment. Low-frequency impedance studies and polarization curves confirmed the formation of a more effective protective film on the steel surface in model solutions of the studied phosphate-based anti-corrosion pigments in comparison with conventional zinc phosphate.

Given the insufficient environmental friendliness of most phosphates, especially zinc, the "green" direction of development of modern technologies is gaining more and more popularity, aimed at developing environmentally safe technological solutions and rational use of natural resources. There are a number of studies that indicate the prospects of using natural materials of plant origin in the manufacture of paint and varnish coatings [6–9]. The work [6] notes the steady and relevant direction of creating new bio-coatings using natural materials, including those of plant origin, such as oils, wood resources, including lignin, cellulose and hemicellulose, to replace petroleum-based materials. However, attention was focused on the noticeable lack of extensive research on the use of lignocellulosic materials for the production of bio-soluble anti-corrosion coatings.

The promising use of bio-resources is also noted in the work [7]. It is noted that the addition of bio-fillers of natural origin has a positive effect on the performance characteristics of epoxy composites due to the intermolecular interaction between natural plant fibers, epoxy resin and hardener. Promising components of bio-coatings can be cashew nut waste, hemp fibers, banana peel, coconut shell powder, rice husk, etc. At the same time, as the authors note, there are not enough experimental, theoretical and model studies to predict the serviceability.

The main factor in the use of natural materials is their extractive part, which demonstrates inhibitory properties in relation to the corrosion destruction of steel. In the work [8], it was emphasized that an important aspect of the stability of paint and varnish coatings is the presence of tannins in plant waste, which contribute to a significant increase in the protective effect of the paint and varnish system, if they are in the primer layer of the coating. Thus, it was shown that a wide range of organic compounds related to tannins are present in various plant raw materials. For example, representatives of tannins procyanidins, catechins and epicatechins, gallates, quercetin and their derivatives were found in the walnut shell. The presence of a significant amount of polyphenolic compounds, representatives of tannins, in the shells of pine nuts, almonds, and walnuts is also confirmed by studies presented in [9].

Among the plant materials studied in paint and varnish coatings, walnut shells are popular. Thus, the authors of the work [10] encapsulated synthesized mesoporous carbon nanospheres with walnut extract, which was confirmed by the authors by thermogravimetric analysis and infrared spectroscopy with Fourier transform. As noted in this study, the protective ability of the epoxy coating in the presence of spheres encapsulated with green walnut shell extract was improved several times.

Since plant extractives play an important role in inhibiting corrosion processes, it is natural that an important issue is the regulation of their extractivity. Thus, according to the results of studies [11], the increase in extractivity, and therefore the anti-corrosion properties of walnut shells, can be significantly increased by ultrasonic treatment of shell powder in solution together with additives of inorganic origin, for example, zinc phosphate ($Zn(H_2PO_4)_2$).

Therefore, a promising direction for increasing the corrosion resistance of paint and varnish coatings is the development of mixtures of known inorganic pigments, such as phosphates, with materials of plant origin – walnut shells. Since aluminum phosphate ($AlPO_4$), unlike zinc phosphate, is more environmentally friendly, optimization of the composition based on it and walnut shells for introduction into anti-corrosion paint and varnish coatings is a promising and relevant task.

The aim of research is to optimize the composition based on walnut shells and anti-corrosion filler aluminum phosphate to increase the corrosion resistance of water-based paint and varnish coatings. To achieve this aim, it is necessary to solve the following objectives:

– to study the effect of the composition of the mixture of walnut shell powder and aluminum phosphate pigment on the corrosion rate of steel in model solutions obtained by extracting components according to the synthesized experimental plan;
– to obtain an adequate mathematical model of the dependence of the corrosion rate on the composition;
– to find the optimal composition and experimentally verify the obtained result.

2. Materials and Methods

2.1. The object and hypothesis of research

The object of research is the anti-corrosion properties of mixtures of walnut shell powder and aluminum phosphate ($AlPO_4$).

Research hypothesis: the inclusion of walnut shell powder together with aluminum phosphate in the composition of water-based paint and varnish coatings will help reduce the corrosion rate of steel. The optimal composition based on these components can be determined using mathematical modeling and experimental analysis, which will ensure the minimum corrosion rate.

During the research, it was assumed that the effectiveness of aluminum phosphate traditionally used in paint and varnish coatings can be increased by using it in a mixture with a vegetable filler containing water-soluble tannins. The formation of insoluble protective chelate complexes of tannins with Al^{3+} ions should reduce the corrosion rate on the steel surface. The search for a mathematical model and the solution of the optimization problem were performed taking into account the assumption that the dependence of the steel corrosion rate on the content and ratio of components in the mixture is nonlinear.

2.2. Materials and methods of research

Walnut shell powder (WP) with a dispersion of 75 μm was purchased from Lingshou Fengfeng Mining Products Processing Factory (PRC), aluminum phosphate $AlPO_4$ (AP) – from Shanghai Macklin Biochemical Technology Co., Ltd (PRC).

WP and AP extracts were prepared by ultrasonic treatment of an aqueous suspension, which included finely ground walnut shell powder and/or aluminum phosphate powder and water in various ratios. Ultrasonic treatment of suspensions was carried out using an ultrasonic homogenizer WH003 (JiagyinWeiheng Industrial Technology Co, China, 220 V/50 Hz) for a total time of 10 minutes according to the scheme, according to which every five seconds there was treatment with an ultrasonic frequency of 20 kHz, at a power of 1200W, and the solution "rested" for the next five seconds. After ultrasonic treatment, the suspension was filtered.

The corrosion rate was determined by the massometric method by exposing steel samples to extracts for 120 hours (5 days). The change in the mass of the samples was recorded on an electronic balance Sartorius BSA124S (Germany).

The corrosion rate was studied on samples measuring 40 × 25 × 1 mm from steel grade Q215, which complies with the GBT700_2006 (China) standard [12]. The main components of cold-rolled steel grade Q215 include: carbon (C): ≤ 0.15%, silicon (Si): ≤ 0.35%, manganese (Mn): ≤ 1.20%, phosphorus (P): ≤ 0.045%, sulfur (S): ≤ 0.050% (grade A), contains trace amounts of residual elements such as chromium (Cr), nickel (Ni) and copper (Cu), which usually do not exceed 0.30% each.

Before immersion in solutions, steel samples were cleaned by processing with P-1920 paper (PRC) with a grain size of 18 μm, degreased with ethanol, dried by blowing hot air, and then placed in a desiccator with a moisture absorber. After one day, they were weighed on a balance with an accuracy of 0.0001 g. After corrosion tests, the samples were cleaned of corrosion products for 1–2 minutes in a 5% sodium thiosulfate solution. Further, the sample processing process was identical to the sample preparation process before testing.

According to the results of massimetric measurements, the mass corrosion index K_m^- and the degree of metal corrosion protection were determined according to the formula [13]

$$K_m^- = \frac{m_0 - m_1}{S \cdot \tau}, \tag{1}$$

where K_m^- – the mass corrosion index, g/(m^2 · h); m_0 and m_1 – the masses of the samples before and after testing, g; S – the area of the metal sample, m^2; t – the sample exposure time in the corrosive environment, h.

The research methodology is schematically shown in Fig. 1.

2.3. Modeling and optimization of the mixture composition of walnut powder and aluminum phosphate

In order to study the influence of the composition of the mixture of WP and AP pigment on the anticorrosive properties of the extractive part of water-based paint and varnish coatings, it is advisable to build a second-order polynomial model of the following form

$$K_m^- = a_0 + a_1x_1 + a_2x_2 + a_3x_1^2 + a_4x_2^2 + a_5x_1x_2, \tag{2}$$

where x_1 – the WP mass fraction in the mixture; x_2 – the $AlPO_4$ mass fraction in the mixture; a_j (j = 0...5) – the coefficients of the mathematical model, which are calculated using known methods [14] based on statistical data.

The adequacy of the model was assessed by the values of the F-ratio, the multiple correlation coefficient R and the standard deviation s

$$F = \frac{s_{itg}^2}{s_{ing}^2}, \tag{3}$$

where s_{itg}^2, s_{ing}^2 – the intragroup and intergroup variances, respectively;

$$R = \sqrt{a_1 r_{K_m^- x_1} + a_2 r_{K_m^- x_2} + \cdots}, \tag{4}$$

where a_j (j = 0...5) – the coefficients of equation (2); $r_{K_m^- x_j}$ – the sample correlation coefficient;

$$\sigma = \sqrt{\frac{1}{n-1}\sum (K_{m\,\exp}^- - K_{m\,calc}^-)^2}, \tag{5}$$

where $K_{m\,\exp}^-$, $K_{m\,calc}^-$ – the experimental and calculated values of the mass corrosion index, respectively; n is the number of experiments.

The significance of the coefficients of the mathematical model (2) was assessed by the Student test by comparing the calculated t_{jp} and critical t_{cr} values of the criterion at a significance level of q = 0.01

$$t_{jp} > t_{cr}, \tag{6}$$

where t_{jcr} is calculated for the coefficient of equation (2) by the formula

$$t_{jp} = \frac{|a_j|}{s_{a_j}}, \tag{7}$$

where s_{aj} – the standard deviation of the coefficient.

The problem of optimizing the composition of the mixture of WP and AP powder is formulated as follows.

Let the effectiveness of anti-corrosion protection of the metal be estimated by the function (2), and the variables are subject to restrictions

$$0 \le x_i \le 1, \sum x_i < 1 \left(i = 1{,}2\right). \tag{8}$$

where x_i (i = 1, 2) – the mass fraction of the WP and AP shell, respectively, in the mixture.

It is necessary to find such a point x^{opt} = (x_1^{opt}, x_2^{opt}), which will provide the minimum value of the function (2).

The optimization of the objective function was performed using the OPTIMIZ-M computing module [15].

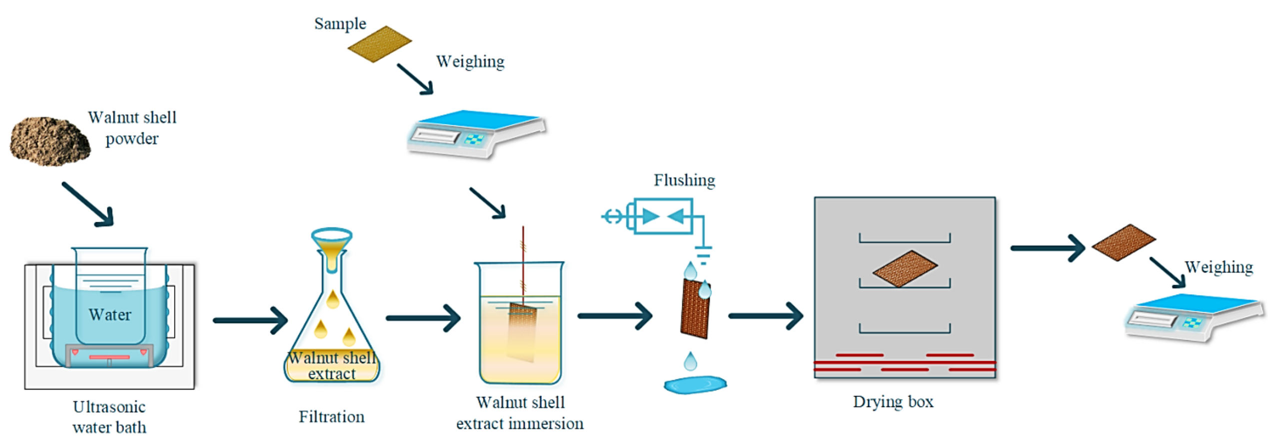

Fig. 1. Graphical representation of the methodology for studying the anticorrosive properties of mixtures of walnut shell powder and pigment

3. Results and Discussion

The content of the mixture components varied from 0 to 1 in increments of 0.2, taking into account the constraints (8). The number of candidate points for the experimental design was calculated as the number of all combinations of different values of the mass fraction of the mixture components by 2

$$A_n^2 = \frac{n!}{(n-2)!}. \quad (9)$$

Taking into account the constraints imposed on the mixture components, the experimental design included the mixture variants that satisfy the condition (8). Table 1 shows the values of the content of the mixture components in coded form.

For each point of the plan, 3 parallel experiments were conducted. The experiment was conducted in the laboratory of Ningbo Xin'an Coatings Co., Ltd. (PRC, Ningbo). The effect of extracts obtained in solutions with 5% and 15% concentrations of powder additive mixtures on the corrosion behavior of steel was studied.

The results of the experiment are presented in Table 2.

Table 1

Experiment design

Mixture component	Experiment number					
	1	2	3	4	5	6
x_1	0	0.2	0.4	0.6	0.8	1
x_2	1	0.8	0.6	0.4	0.2	0

Note: x_1 is the mass fraction of walnut shell powder, and x_2 is the mass fraction of aluminum phosphate

Table 2

Experimental values of the mass index of corrosion rate, g/(m^2 · h)

Experiment number					
1	2	3	4	5	6
5% solution					
0.0361	0.0569	0.0819	0.0653	0.1042	0.1181
15% solution					
0.0570	0.0900	0.1070	0.1170	0.1180	0.1480

The mathematical model in its natural form after processing the experimental results and rejecting insignificant coefficients according to the Student's criterion took the form

$$K_m^- = 0.115 + 4.1\cdot 10^{-4}x_1 - 2.2\cdot 10^{-2}x_2 + 1.4\cdot 10^{-3}x_2^2 + 9.4\cdot 10^{-4}x_1x_2. \quad (10)$$

The value of the multiple correlation coefficient $R = 0.92$ indicates a fairly close relationship between the values of the mass index of the corrosion rate and the composition of the mixture. The difference between the experimental $K_{m\,\exp}^-$ and the calculated $K_{m\,calc}^-$ values of the mass index of the corrosion rate, as well as other characteristics of the model given in Table 3, confirm the adequacy of the model.

The optimal values of the components in the WP and AP mixture, found using the OPTIMIZ-M program: $x_1 = 0.11$; $x_2 = 0.89$. The developed composition of the mixture provides the minimum value of the mass index of the corrosion rate $K_{m\,\exp}^- = 0.02002$ g/(m^2 · h).

The dependence of the mass index of the corrosion rate on the content of the mixture components at the optimal point (x_1^{opt}, x_2^{opt}) is shown in Fig. 2, 3. The fixed values of the mixture components correspond to the optimal values – x_1^{opt} and x_2^{opt}, respectively.

Table 3

Comparison of experimental and calculated values of the mass index of the corrosion rate

Mass index of corrosion rate, g/m^2·h	Experiment number						Model characteristics
	1	2	3	4	5	6	
Extract obtained in a 5% solution (g/100 g)							
$K_{m\,\exp}^-$	0.0361	0.0569	0.0819	0.0653	0.1042	0.1181	*F*-ratio: 6.38
$K_{m\,calc}^-$	0.0405	0.0541	0.0686	0.0839	0.1000	0.1171	Multiple correlation coefficient: 0.92
Δ	−0.0044	0.0028	0.0134	−0.0186	0.0041	0.0010	
Extract obtained in a 15% solution (g/100 g)							
$K_{m\,\exp}^-$	0.0570	0.0900	0.1070	0.1170	0.1180	0.1480	Standard deviation: 1.87·10^{-3}
$K_{m\,calc}^-$	0.0540	0.0880	0.1010	0.1210	0.1170	0.1150	
Δ	0.0030	0.0020	0.0060	−0.0040	0.0010	0.0330	

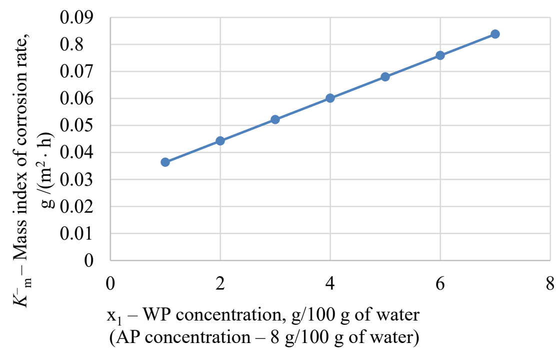

Fig. 2. Dependence of the value of the mass index of steel corrosion rate K_m^- on the concentration (g/100 g of water) of WP powder x_1 at a fixed value of the concentration of AP pigment x_2

Analysis of the results shows that at a constant concentration of phosphate pigment, the corrosion rate increases with increasing walnut shell powder concentration. The minimum corrosion rate is observed at a concentration of 1 g/100 g of water. The increase in the corrosion rate with increasing walnut shell powder content is explained primarily by the chemical composition of its extract. Aluminum phosphate $AlPO_4$ due to strong ionic bonds between Al^{3+} and PO_4^{3-} in the crystal lattice is almost insoluble in water (0.001 g/l). Al^{3+} ions in water form the ion $[Al(H_2O)_6]^{3+}$, which is further hydrolyzed with acidification of the medium due to the release of H^+ ions. An increase in the solubility of aluminum phosphate can be achieved by acidifying the medium. On the other hand, tannins contained [8, 9] in walnut shells always contribute to the acidification of the aqueous medium. With an increase in the proportion of walnut powder in the solution, the acidification of the solution will occur more intensively, which will automatically lead to an increase in the solubility of aluminum phosphate. That is, the acidity of the solution will increase due to an increase in tannins extracted from walnut shell powder and acidification by Al^{3+} ions. Thus, an increase in the proportion of walnut shells at constant aluminum phosphate content will contribute to an increase in the corrosion rate of steel due to an increase in the contribution to the overall corrosion process of the rate of the cathodic hydrogen depolarization reaction, which was confirmed in the results of the research (Fig. 2). It is well known that tannins are known for their chelating properties and can chelate, i. e. bind various metal ions, including Al^{3+} ions, into insoluble compounds. In [16] it was shown that Al^{3+} ions form complexes with epicatechin polymer (or tannin) in a weakly acidic medium (pH 5–7). The authors of [17] established that between high molecular weight polyphenols and Al^{3+} ions tannin complexes are formed with different stability, which depended on the pH of the solution. It was shown that at pH 6 the conditional stability constant (β) was ~ 1 · 10^{23}, and at pH 4 it significantly decreased to $\beta \sim 1 \cdot 10^5$, which indicates a significant decrease in the stability of tannin complexes with a decrease in pH. Therefore, it can be assumed that at optimal ratios, under conditions of a relatively weak increase in pH acidity, tannins will be capable of forming poorly soluble chelate-type compounds, which will form a protective film on the surface. Fig. 3 shows the level of the corrosion process at a constant concentration of tannins due to walnut shells (1 g/10 g of water) with an increase in the concentration of aluminum phosphate. The results presented in Fig. 3 may indicate a high probability of the formation of a protective film within certain limits of the ratios of Al^{3+} ions and tannins. Within the limits of increasing the content of aluminum phosphate from 1 g to 7.5 g per 100 g of water, a decrease in the corrosion rate was observed. Probably, within these limits of concentrations, tannins form more stable complex compounds that form a protective film on the steel surface. However, tannin complexes are more stable only within certain limits (pH 4–6), therefore, a further increase in aluminum phosphate in the solution contributes to a decrease in pH to the limits when more soluble complexes with a low protective ability of the film formed on the surface begin to form (Fig. 3).

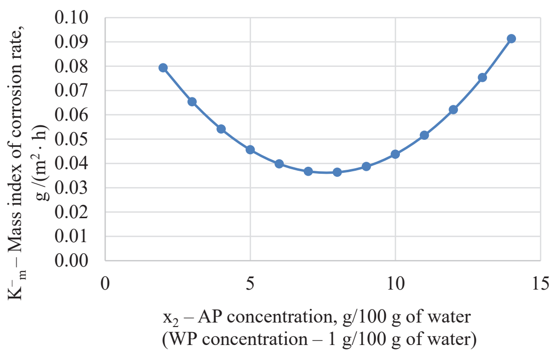

Fig. 3. Dependence of the value of the mass index of steel corrosion rate K_m^- on the concentration (g/100 g of water) of the pigment x_2 – aluminum phosphate – at a fixed value of the concentration of walnut shell powder x_1

The response surface area of the mass index of steel corrosion rate in aluminum phosphate and walnut shell powder extracts depending on the content of the mixture components is shown in Fig. 4.

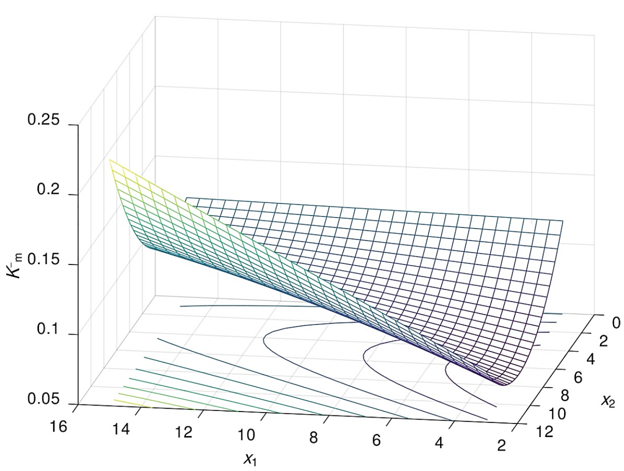

Fig. 4. Surface area of the response of the mass index of corrosion rate depending on the concentration (g/100 g of water) of walnut shell powder – (x_1) and aluminum phosphate pigment (x_2)

The analysis of the results shows that it is rational to prepare mixtures based on walnut shell powder and aluminum phosphate at a component ratio of about 8:1, which ensures the minimum rate of the corrosion process. Experimental verification of the composition of the mixture showed that at optimal component ratios the steel corrosion rate is 0.018 g/(m^2 · h), which practically corresponds to the value obtained by calculation (0.020 g/(m^2 · h)).

The limitation of this research is that the results were obtained only for walnut shell as a component of an anti-corrosion composition based on aluminum phosphate. Other plant materials, such as bamboo powder, will have a different composition of the extractive part, which will affect the level of steel corrosion rate. Prospects for further research are to study the mechanism of anti-corrosion action of walnut shell powder with other traditionally used pigments, which will allow the development of more environmentally friendly paint and varnish compositions.

4. Conclusions

The possibility of increasing the corrosion resistance of water-based paint coatings was studied by using a mixture of environmentally common components in the compositions, namely aluminum phosphate pigment and walnut shell powder. The studies were conducted according to the synthesized optimal experiment plan. The experimental data were used to develop an adequate mathematical model that describes the relationship between the corrosion rate and the composition of the mixture of aluminum phosphate and finely ground walnut shell powder. Using the author's mathematical optimization program OPTIMIZ-M, the values of the mass fractions of the components in the mixture of walnut shell powder and aluminum phosphate (0.11 and 0.89, respectively) were found, which provide the minimum value of the mass index of the corrosion rate of 0.020 g / (m^2 · h). Experimental verification of the obtained results showed that the optimal ratio of components in a mixture based on phosphate and walnut shell is 8:1. This ratio reflects the conditions for the formation of poorly soluble complexes of aluminum ions extracted from aluminum phosphate pigment and tannin compounds from walnut shell extract. A mixture of aluminum phosphate and walnut shell can be successfully used in the production of water-based paint and varnish coatings, which will increase their environmental friendliness without losing anti-corrosion properties.

The obtained results have practical significance, since they allow demonstrating ways to increase the efficiency of pigments and fillers in paint and varnish coatings, primarily by creating mixtures using plant waste, which will allow obtaining more environmentally friendly paint and varnish compositions.

Conflict of interest

The authors declare that they have no conflict of interest regarding this research, including financial, personal, authorship or other, that could influence the research and its results presented in this article.

Financing

The research was performed without financial support.

Data availability

The manuscript has no associated data.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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