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Original Empirical Research

Optimization of the Paintwork Material Modified by Metal Catalyser Additive

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Abstract
Introduction. Current trends in the paint and varnish materials industry are embracing environmental friendliness and versatility lending wooden products good aesthetic and protective properties. The structure of wood, as a natural material, is constantly undergoing intensive and progressive processes of oxidative degradation under environmental conditions affecting the strength of wood and causes significant structural changes. Therefore the interest in improving the durability of paint coatings under the influence of environmental factors on their performance justifies the intensification of research in the development of new effective solutions. One of the effective ways to prevent the destruction of the wood structure is to apply a protective layer of paint and varnish material by chemically modifying the surface and, above all, by introducing siccatives. The introduction of siccatives makes it possible to ensure a uniform drying rate throughout the entire volume and additionally disperse the pigment, which improves the physical and mechanical properties of the paintwork and increases its durability.
The purpose of the research work is to establish the effect of the addition of a metallic catalyst in the form of highly dispersed precipitation waste from an electric arc furnace on the physical and mechanical properties of paint and varnish materials.
Materials and Methods. The initial components for obtaining oil paint compositions (paintwork material) were used in the experiments as: binder-natural olifa, pigment-ochre, fine aggregate-chalk. To the intensification of the drying process, the addition of metal catalyser, which is a highly dispersed waste of deposition from electric arc furnace, was introduced. The granulometric composition of chalk was evaluated using scanning electron microscopy, and dust using a microsizer 201c laser analyzer.
Results. According to the results of the optimisation, regression equations represented as a polynomial of the second degree and the optimal material composition of the paint material were obtained. In order to solve the problem of drying, a metal catalyser was added to the optimal composition in the amount of 0.05 % of the binder weight. A comparison of the obtained results of regulatory tests of the physical and mechanical properties of the two formulations, the control (without additives) and the modified with the addition of a metal catalyst in the form of dust, indicate the prospects of its use as a siccative.
Discussion and Conclusion. The introduction of a siccative into the oil-based paint and varnish material in the form of a by-product of highly dispersed precipitation waste from an electric furnace accelerated the polymerization process and improved the physical and mechanical properties of the modified composition in comparison with the control one. Improving the physical and mechanical characteristics of oil paint will increase the resistance of coatings to environmental factors and thus increase its durability.

Keywords: paintwork material, adhesion, nominal viscosity, metal catalyser, fine aggregate, binder

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Оригинальное эмпирическое исследование

Оптимизация лакокрасочного материала для покрытия древесины с добавкой металлического катализатора

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Аннотация
Введение. Современные тенденции в отрасли производства лакокрасочных материалов идут в направлении экологичности и многофункциональности придавая деревянным изделиям хорошие эстетические и защитные свойства. Структура древесины, как натурального материала постоянно подвергается интенсивным и прогрессирующим процессам окислительного разрушения в условиях воздействия окружающей среды, что влияет на прочность древесины и вызывает значительные структурные изменения. В этой связи интерес к улучшению стойкости лакокрасочных покрытий при воздействии факторов окружающей среды на их эксплуатационные характеристики, оправдывает активизацию исследований в разработке новых эффективных решений. Одним из эффективных способов предотвращения разрушения структуры древесины является нанесение защитного слоя лакокрасочного материала путем его химической модификации поверхности и прежде всего за счет введения сиккативов. Введение сиккативов позволяет обеспечить равномерную скорость высыхания по всему объему и дополнительно диспергировать пигмент, что улучшает физико-механические свойства лакокрасочного покрытия и повышает его долговечность.

Цель научно-исследовательской работы — установить влияние добавки металлического катализатора в виде высокодисперсного отхода осаждения от электродуговой печи на физико-механические свойств лакокрасочного материала.

Материалы и методы. В качестве исходных компонентов для получения масляных составов красок при проведении экспериментов применяли: связующее вещество — натуральная олифа, пигмент — охра, наполнитель — мел. Для ускорения процесса высыхания добавляли металлический катализатор, являющийся высокодисперсным отходом осаждения от электродуговой печи (далее — пыль). Гранулометрический состав мела оценивали с помощью сканирующей электронной микроскопии, а пыли — с помощью лазерного анализатора Microsizer 201c.

Результаты исследования. По результатам оптимизации были получены уравнения регрессии, представленные в виде полинома второй степени и оптимальный вещественный состав лакокрасочного материала. Для решения проблемы высыхания в оптимальный разработанный состав масляной краски вводили добавку металлического катализатора в количестве 0,05 % от массы связующего.

Сравнение полученных результатов нормативных испытаний физико-механических свойств двух составов контрольного (без добавки) и модифицированного с добавкой металлического катализатора в виде пыли говорят о перспективности её применения в качестве сиккатива.

Обсуждение и заключение. Введение в лакокрасочный материал на масляной основе сиккатива в виде побочного продукта высокодисперсного отхода осаждения от электродуговой печи ускорило процесс полимеризации и улучшило физико-механические свойства модифицированного состава в сравнении с контрольным. Улучшение физико-механических характеристик масляной краски позволит повысить стойкость покрытий к воздействию факторов окружающей среды и таким образом повысить его долговечность.

Ключевые слова: адгезия, условная вязкость, лакокрасочный материал, металлический катализатор, наполнитель, связующее

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Introduction. The growing consumption in the global market of paint and varnish materials can be put down to the trend of "green" buildings in civil engineering generating a soaring demand for effective "green" paint and varnish materials. Oil and paint industry is moving in two relevant areas of great practical interest, i.e., bioresources and renewables. In fact, as the industry is putting more emphasis on ecological and multifunctional synthetic fillers [1, 2] whose production does not normally take into account the characteristics of environmental sustainability [3], scientific research emphasizes the use of natural additives in coatings [4, 5]. Scientists have thus recently investigated the results of the use of linseed oil [6], pigments obtained from wood waste [7], microbial staining [8], pigments extracted from fungi [9] and cellulose fibers [10] in wood coatings. The paint and varnish material lending wood products specific aesthetic effects with new pigments [11] and distinctive gloss indicators [12] is indicative of the trend gaining momentum in the market of protective coatings for wood. On the one hand, while using innovative pigments, unique aesthetic effects should be provided making them weather-resistant without compromising the protective qualities of the organic coating [13]. On the other hand, the combination of wood paints with diverse types of pigments might lead to grave problems potentially reducing the protective effectiveness of the organic coating, introducing a gap in the polymer matrix and showing a low level of inherent durability of the pigment [14, 15].

The use of oily formulations shields wooden products and structures from occasional moisture exposures causing rotting accompanied by a loss of integrity and load-bearing capacity. The film-forming substance in oil formulations are drying oils, which, according to their content of polymers and processed vegetable oils, are classed into:

1) natural drying oils;
2) semi-natural drying oils;
3) artificial drying oils.

Natural drying oils are obtained by heating linseed, hemp or other vegetable oils to an average of 150 °C. At the same time, the addition of solidification accelerators (siccatives) is introduced in an amount of 2–4%. Due to this treatment, drying oil takes on the property of quickly "drying out" in the air forming an elastic film. Oil-based colorful formulations are obtained by meticulously grinding drying oil with pigments insoluble in oils in machines. While grinding colourful formulations, homogeneous suspensions should be obtained where each particle of pigment or filler has a shell of binder adsorbed on the surface of the particles. The wettability of the pigments by the binder, the strength of the resulting shells and thereby the properties of the paint compositions depend on those of both the pigment and the wetted liquid. Pigments can be hydrophilic (iron meerkat, ochre) and hydrophobic (graphite, soot, lead whitewash). When hydrophilic pigments are mixed with oil binders that do not contain surfactants, there is only a mechanical mix with reduced painting properties. The presence of surfactants in the composition enhances the wettability of the pigment with oils. One of the drawbacks of drying oils is the relatively long drying time. Despite certain disadvantages [16] the use of metal catalysts can reduce the drying time considerably.

The wettability scheme of the surfactant pigment in the presence of a metal catalyst in Fig. 1 is the following. Surfactants are attached by a polar group to the surface of the pigment and form a hydrophobic shell that is well wetted with an oil binder, and the inner surface of the shell binds firmly to the pigment and the metal catalyst.

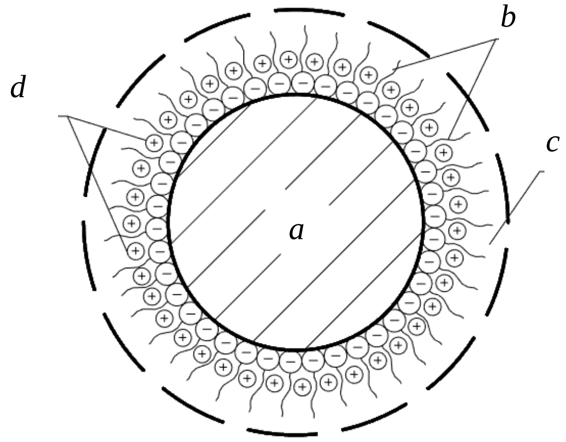


Fig. 1. Scheme of wettability of surfactant pigment in the presence of a metal catalyst:
a — pigment; *b* — oriented molecules of the surfactant pigment; *c* — oil binder; *d* — metal catalyzer

The objective of the research is to identify the effect of adding a metal catalyst in the form of highly dispersed precipitation waste from an electric arc furnace on the physical and mechanical properties of oil paint.

Materials and Methods. The initial components for obtaining oil compositions of paints during the experiments were as follows: the binder is natural drying oil, the pigment is ochre, the filler is chalk (CaO — 50.90%; PP — 41.50%) with a specific surface area of 420-436.5 m^2/kg. Experimental studies carried out by means of scanning electron microscopy (Fig. 2) show that chalk particles are mostly characterized by a rounded shape ranging from 2 to 20 microns in size.

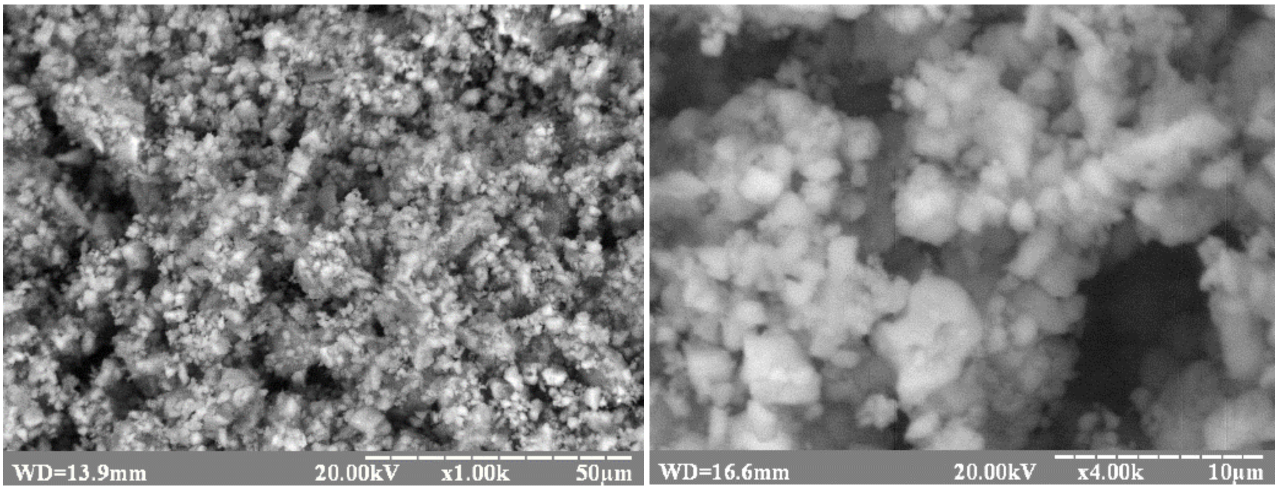


Fig. 2. Scanning electron microscopy (SEM)

In order to accelerate drying, an additive of a metal catalyst was introduced, which is a highly dispersed precipitation waste from an electric arc furnace. The chemical composition of the metal catalyst are mainly metal oxides (ZnO — 46.7%; Fe_2O_3 — 32.4%, etc.). The granulometric composition of the dust performed with a Microsizer 201c laser analyzer is shown in Fig. 3.

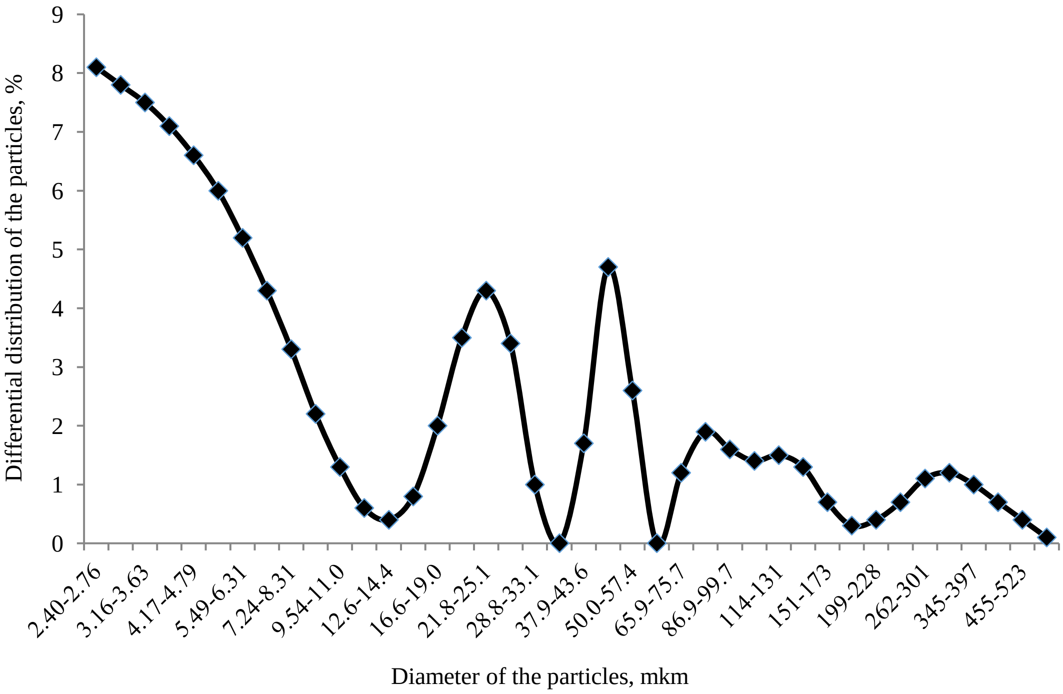


Fig. 3. Granulometric composition of the metal catalyst

The fundamental properties of oil paints depend on the amount and type of pigment, and the amount of binder varies a lot. According to the results of the literature review, it is assumed that the amount of the binder lies in the range of 20–40%, the filler — 15–40% and the pigment — 15–35%.

Optimization. The development of the composition of an effective paint and varnish material for wood coating was determined with the optimal ratio between the components. The experimental data was processed by means of the least squares method in matrix form. The variable factors were: x_1 — pigment content, %; x_2 — filler content, %. The function of the output parameters were the following: y_1 — adhesion to the paintwork, MPa; and y_2 — conditional viscosity, C. The values of the variation factors and their physical value are in Table 1.

Table 1

Significance of variation factors in a complete factor experiment (CFE) 2^k

№	Factor code	Physical value of the factor	Measurement unit	Variation range	Factor levels		
					–1	0	+1
1	x_1	pigment content	%	± 5	15	25	35
2	x_2	filler content	%	± 10	17.5	27.5	37.5

Identifying the drying time. A thin layer of paint and varnish material was applied to the prepared 70 ×150 mm steel plates without leaving any gaps. With a light touch of the fingers, the stickiness was gone, and only after that were 0.5 g of Ballotini glass beads (with a fraction from 100 to 355 microns) poured onto the surface of the plates in one layer from a height of 50 mm to an area with a diameter of 22 mm. After 60 seconds, the plate was tilted at an angle of about 20 ° in relation to the horizontal and swept away with a soft brush. The degree of drying is achieved if all the Ballotini balls are removed with no damage to the surface layer. The time corresponding to the degree of drying is recorded.

Identifying the conditional viscosity based on the B3-246 viscometer. The paint and varnish material with a volume of 150 cm^3 was filtered through a sieve to eliminate foreign inclusions. Prior to testing on the B3-246 viscometer, the nozzle was thoroughly cleaned with a solvent. The nozzle of the tank was closed with a rod and the resulting composition was poured to a horizontal mark on its inner surface. A glass measuring cup was placed under the nozzle of the viscometer. After the paintwork had been poured, the rod was lifted. As the test paint and varnish material were coming out of the nozzle, a stopwatch was turned on and the conditional viscosity was identified according to the time.

Identifying the mass fraction of non-volatile substances. For the test, a metal cup with an inner diameter of the bottom (75 ± 5) mm and a side height of at least 5 mm was used. By weighing, the mass of an empty dry cup m_0 was identified using an analytical balance with an accuracy of 1 mg. A sample of the paint and varnish material was then added and the mass in the cup (m_1) was identified, while evenly distributing it over the diameter of the cup. After the mass had been identified, the samples with the cup were placed in a drying cabinet with a temperature of 105-110°C and dried to a constant mass. Having been dried and cooled at room temperature, the mass of the cup with the dried residue (m^2) was identified yet again. The mass fraction of non-volatile substances *HB*, %, was calculated using the formula:

$$\mathrm{HB} = \frac{(m_2 - m_0)}{(m_1 - m_0)} 100\%, \quad (1)$$

where m_2 is the mass of the cup with the remainder, g; m_0 is the mass of the empty cup, g; m_1 is the mass of the cup with the sample prior to the tests, g.

Identifying the adhesion of the paintwork by the strength of the tear. A paint and varnish material was applied to the selected metal prepared surface, which was pre-primed in several layers. After the paintwork had been cured, metal discs were glued to the samples, and the remaining glue was removed and held in such a manner until it solidified completely. Next, the samples were fixed in an adhesive device, and the metal discs glued to the samples were pivotally connected to the adhesive gripper as shown in Fig. 4. According to a visual assessment of the adhesive strength R_{ad} = 1.8 MPa, the zone and nature of the disc separation from the metal base were recorded.

Fig. 4. Adhesive meter ONYX-1.AP.020

Identifying the adhesion with the method of lattice incisions. The optimal oil composition was applied in two layers to the prepared plates measuring 60×150×1 mm. The tests were conducted with a razor blade and at least six parallel incisions were made along a ruler with a length of at least 20 mm at a distance of 1-3 mm from each other. After the incisions had been made and the loose pieces of the coating had been removed, a soft brush was applied diagonally across the surface of the grate.

Research Results. According to the results of the experiment, regression equations (2) and (3) were obtained presented as a polynomial of the second degree. Regression equations processed using the least squares method were subjected to statistical analysis based on the estimates of variances. The calculation of the regression coefficients and statistical criteria is shown in Tables 2, 3. A graphical interpretation of the obtained equations is shown in Fig. 5, 6.

$$y_1(x_1,x_2)=2{,}09–0{,}235\cdot x_1+0{,}18\cdot x_2–0{,}55\cdot x_1\cdot x_2–0{,}335\cdot x_1^2+0{,}02\cdot x_2^2 \quad (2)$$

$$y_2(x_1,x_2)=15{,}59–3{,}94\cdot x_1+2{,}94\cdot x_2+0{,}278\cdot x_1\cdot x_2+0{,}278\cdot x_1^2–0{,}667\cdot x_2^2 \quad (3)$$

Table 2

Calculated coefficients of the equations

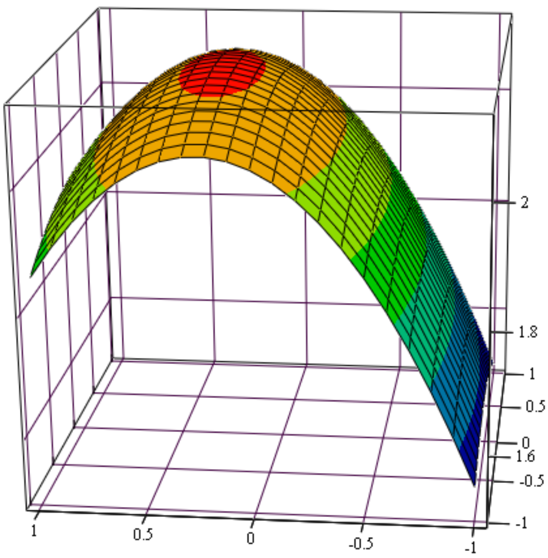
Name of the output parameter of the equation	Coefficients of the equations					
	b_0	b_1	b_2	b_3	b_4	b_5
y_1	2.09	–0.235	0.18	–0.55	–0.335	0.02*
y_2	15.59	–3.944	2.94	0.278*	0.278*	–0.67*

* — insignificant coefficients

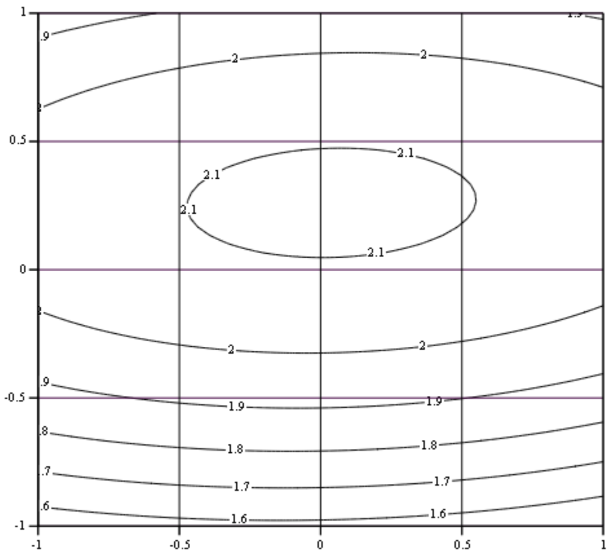
Table 3

Statistical optimization criteria

Name of the output parameter of the equation	Statistical criteria			
	F	D^2_0	S_0	ζ
y_1	6.242	4.107	0.064	0.111
y_2	3.035	0.87	0.933	1.618



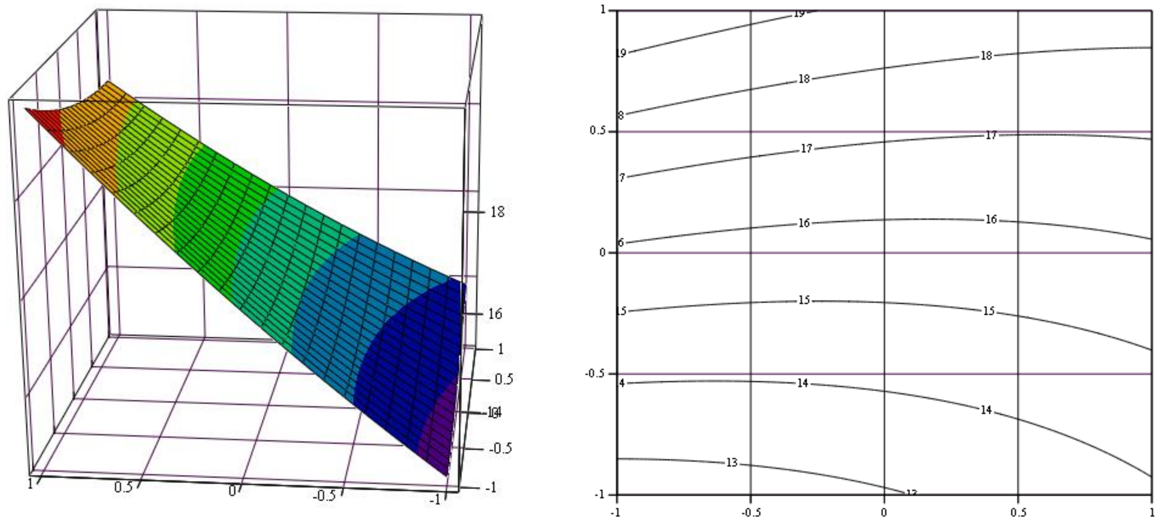
a)



b)

Fig. 5. Dependence of the adhesion on the amount of the pigment x_1 and the filler x_2: *a* — in space; *b* — on the plane

An analysis of the results of the obtained model in the form of regression equations shows that the coefficient b_1 with the factor x_1 (the amount of the pigment) has a negative impact on both of the output parameters (y_1; y_2). Furthermore, the amount of the pigment has a greater effect on the conditional viscosity of the paintwork (b_1 = –3.944) than on the adhesion to the paintwork (b_1 = –0.235). At the same time, a positive value of the b_2 coefficient at the factor x_2 (the amount of the filler) indicates that an increase in the filler concentration results in a positive effect on both adhesion to the paintwork (b_2 = 0.18) and the conditional viscosity (b_2 = 2.944). The combined interaction of these factors (x_1; x_2) is insignificant, but negatively impacts adhesion (b_3 = –0.55) in the first case and positively impacts the conditional viscosity (b_3 = 0.278) in the second one. The remaining coefficients can be neglected as they are insignificant in terms of the conditional viscosity (b_4; b_5) and adhesion (b_5), since it is less than the root-mean-square error in identifying the coefficients.



a) *b)*

Fig. 6. Dependence of the conditional viscosity on the amount of the pigment x_1 and the filler x_2: *a* — in space; *b* — on the plane

In general, based on the inequality $F < q_F$, with a probability of 0.95, the suggested models are adequate and be employed to describe the optimization process within 0.5. As a result, the optimal composition of the paint and varnish material based on drying oil and pigment in the form of ochre was identified. The material composition of the paint and varnish material is shown in Table 4.

Table 4

Optimal material composition of oil paint

Name of the composition	Content of the components, %			Index	
	Binder	Pigment (ochre)	Filler (chalk)	adhesion to the coating, MPa	conditional viscosity, sec
С№ 3	45	17.5	37.5	1.8	120

In order to address the problem of drying in the optimal composition of the paint and varnish material (Table 4) a metal catalyst additive was added in an amount of 0.05% by the weight of the binder. The use of this additive as a siccative was due to its necessary influence in the two directions. The first one was as a dispersing agent of pigments, and the other one was to improve oxygen penetration into the film volume and thereby to accelerate the drying of the coating. This should cause a reduction in the formation of a matte effect and allow one to maintain a high gloss surface. The results of comparing the physical and mechanical properties of the compositions with the addition of a metal catalyst (№4) and without additives are in Table 5.

Table 5

Results of comparison of the physical and mechanical properties of the oil paint compositions

№	Name of the index	Name of the composition	
		Control (С№ 3)	Operating (С№ 4)
1	Conditional viscosity according to the VZ-246 type viscometer with a nozzle diameter of 4 mm at a temperature of (20,0± ± 0,5) ºC, sec	120	110
2	Drying time to degree 1, no more, at a temperature of (20 ± 2) ºC, min	20	15.5
3	Mass fraction of non-volatile substances, %	88,5	90,4
4	Adhesion of the coating, points	2	1
5	Adhesion of the coating, MPa	1.8	2.15

Discussion and Conclusion. Based on the optimization results, regression equations were obtained as a polynomial of the second degree and the optimal material composition of the paint and varnish material. In order to address the drying problem, a highly dispersed by-product of deposition from an electric arc furnace was introduced into the optimal composition as a metal catalyst in an amount of 0.05% by the weight of the binder. It was found that the drying time to degree 1 of the operating composition (from № 4) is 22.5% lower compared to the control composition (from № 3), the mass fraction of non-volatile substances is 19.8% lower, and the adhesion of the paint coating is 19.4% higher respectively. The composition of an effective oil paint based on drying oil, ochre, chalk and a siccative additive in the form of a metal catalyst was developed and introduced in order to accelerate the drying and additional dispersion of the pigment, thereby improving the coating's resistance to atmospheric influences. The obtained comparative data from the regulatory tests of the physical and mechanical properties of oil paint using a by-product of deposition from an electric arc furnace are indicative of the prospects of its use as a siccative for paint and varnish materials.

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