of competences corresponding to "modern requirements" and distinguish definitely the key ones and we have to be in a constant searching a competence paradigm in a higher school.

The problems of the modern higher school in the context of a competence approach prompt us to creating new educational standards, to forming and realizing our own historical project of the Kazakhstan higher school modernization.

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OPTIMIZATION OF THE PROCESSES OF DISAGGREGATION OF PIGMENTS AND FILLERS USING THE METHODS OF PROBABILISTIC DETERMINISTIC MODELING

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An important technological characteristic of paintwork materials, which determine the coverage and protective properties of their coatings, is the degree of disaggregation of pigments and fillers. In freshly prepared suspensions of paintwork materials the development of aggregation and disaggregation processes depends both on the surface properties of the solid-phase components themselves and on the qualitative and quantitative composition of cover-formers, solvents and surfactants. To optimize the composition of coatings, it is necessary to know how the set of all these components will affect the dispersion processes, which involves the construction of a generalized mathematical model of the system under study.

We attempted withdrawal of multifactor models of the dispersion process of the pigment of titanium dioxide (R-02), and filler, calcite (M-20), in the coating system on the basis of the varnish PF-060, white spirit, surface-active substances (surfactants) AS (an amine derivative with the number of carbon atoms in the chain 6-8, TU 655-RK 056006434-002-2000) on the basis of the principles of probabilistic and deterministic simulation.

Keywords: dispersion, pigment, surfactants, disaggregation, suspension, average diameter, fraction, modification.

Introduction

An essential factor affecting the effectiveness of protective and decorative properties of paint-and-lacquer coating is the degree of dispersion of the pigments and fillers entering into their compositions. Modern possibilities of dispersing equipment provide the required characteristics, however, at the stage of preparation and storage, due to the thermodynamic instability of highly disperse paint-and-lacquer coating, the processes of aggregation (flocculation) are developed, which degrades the quality of the coatings. Modification of paint and varnish compositions due to use of surface acting agents with inhibitory activity in their formulations, as well as the ability to formpermanent (cohesive, adhesive), decorative, insulating films based on their selective adsorption, seems to be a promising improvement of the coatingperformance characteristics. The mechanism of their physicochemical effects on the interface and the dispersed medium as a whole depends on the nature of the surface acting agents and the contacting phases, as well as their quantitative relationships.

The solid ingredient combining process, which includes a pigment with a liquid polymer medium, is very important in the paint coating manufacture since the product technological and paint properties depend on it, as well as many coating performance properties. The main thing in this process is the pigment interaction with the polymer at the interface between the solid and liquid phases, since its intensity determines the solid particle dispersion in the given medium and the nature of the resulting combined “solid phase-polymer” structures that form the subsequent properties specific for real materials. This was established in Academician P.A.Rebinder’s publications addressing the structure formation and physicochemical mechanics of dispersed systems [1-3] and was further developed by his school staff, A.B.Taubman and S.N.Tolstaya [4-6], who studied pigments interacting with polymers at the phase interface for a wide range of model and real objects. An important technological characteristic exhibited by paint coating materials, which determine the coating structural and mechanical properties (coverage, hardness, strength), is the disaggregation degree specific for pigments and fillers [1].

It was found that the pigment particle dispersion, when combined with the polymer, may both improve and worsen (which has been shown in Figure 1 schematically), in addition, it depends on the chemical nature of both components.

The component combination where the solid particle dispersion in the given polymer medium increases, i.e. the aggregates gradually disintegrate to smaller aggregates or even to primary particles, which is shown in Figure 1 – a), b), c), is considered appropriate. Interacting with one another through the surface-active material interlayers, solid particles form a developed structure where, as in the framework, a secondary structure of the oriented (and hence strengthened) polymer is formed [2].

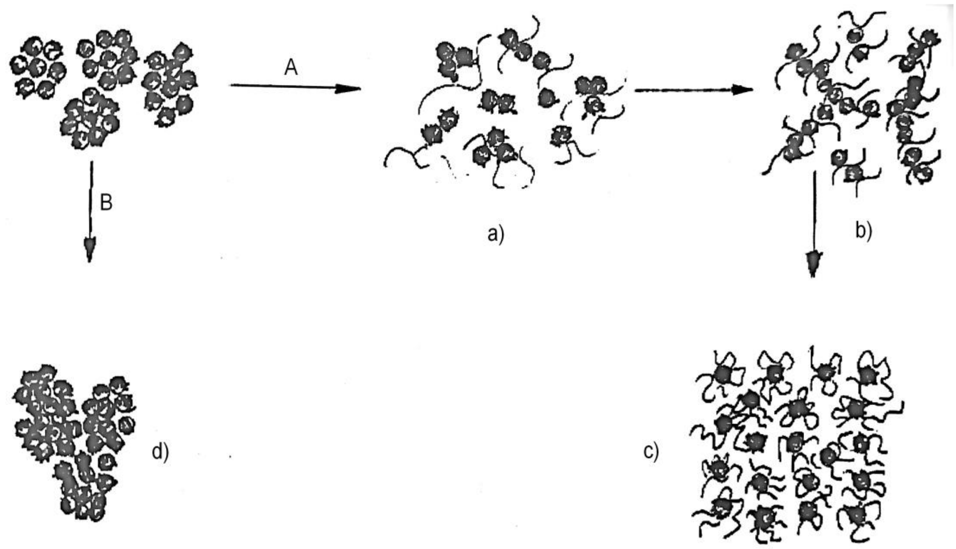

Figure 1 – Change in the pigment dispersion upon its introduction into the polymer medium: A – pigment particle dispersion improvement in comparison with the initial dispersion rate due to natural aggregate peptization: a) separate chain particle formation; b) solid spatial grid formation out of the solid phase particles; c) complete solid particle stabilization. B – pigment dispersion worsening compared with the initial dispersion rate (compact coagulation) – d)

Such combined "solid phase-polymer" structures are called coagulation structures (according to Rebinder); during their formation the system strength increases at the beginning, which is detected by measuring the ultimate statistical shear stress which magnitude is as greater as stronger the solid particle aggregates are peptized in the given medium [7]. Then, after the peptized particles achieve complete stabilization, the strength decreases. Such structures are usually thixotropic, i.e. they are able to recover their physical and mechanical, as well as rheological properties naturally when destructed [3].

For the overwhelming majority of paint coating materials, when combining a pigment with polymer solutions, it is necessary to form coagulation-type structures, since in this case it is possible to obtain materials and coatings based on them having the necessary properties.

However, in many cases the pigment particle peptization process does not occur naturally, on the contrary, formation of larger aggregates can be observed in comparison with the initial particles, i.e. there is a dispersed phase coagulation process in the given medium, as shown in Fig. 1 b, d. The first process occurs under the liquophilicity of the solid phase particle surface against the liquid medium, and the second one occurs under its liophobicity. For example, hydrophobic graphite is well combined with vegetable oils or non-polar rubbers and disperses well in them, whereas hydrophilic inorganic pigments (ZnO, TiO_2, Fe_2O_3, SiO_2) poorly combine with such hydrocarbon media, which can lead to compact solid phase particle coagulation [3]. In systems with weak and medium interaction, the process where the pigment and the filler combine with the polymer must be intensified. Using surface-active substances (SAS), fine regulators of interface interactions and finished material properties proves to be the most effective method.

The SAS effect is based on forming the surface pigment and filler of the adsorption layer, whose properties, in turn, are determined due to the forming its inner and outer parts. The inner adsorption layer part is formed resulting from the polar SAS group interaction with active surface centers, which promotes peptizing the aggregated pigment and filler particles and the formation of coagulation structure elements. The adsorption layer outer part is formed when hydrocarbon SAS molecule radicals are oriented to the polymer medium, increasing the liquophilicity of the pigment particle surface that is combined with it [2].

Polar hydrophilic pigments introduction into non-polar hydrocarbon media, provides for using modifiers with long hydrocarbon chains (hydrophobisators), for example saturated carboxylic acids, amines or their salts, as well as quaternary ammonium salts and other compounds. When it comes to polar dispersion media, SAS whose molecules contain polar groups in the hydrocarbon radical that are lyophilic with respect to the polymer dispersion medium (i.e., various bifunctional compounds) are used as pigment surface modifiers [14-16]. This is applicable to all paint coating material types containing polymer solutions as a binder in organic solvents (or in water for water-soluble polymers), water-dispersed binders. Developed and used in recent years polymeric SAS can be even more effective than their low molecular weight analogues due to their high affinity for the polymer medium.

However, the SAS action in paint coating systems is subject to certain physicochemical laws that must be observed for their effective application. An important role in this is played by the polar SAS group nature and the hydrocarbon radical structure specific for their molecules, as well as the polymer chemical nature and the presence of polar functional groups in its molecules. In the paint coating material suspensions, the development of aggregation and disaggregation processes depends both on the surface properties shown by the solid-phase particles and on the quantitative-qualitative composition of the film-forming agents, solvents and surface-active additives.

An urgent task is to improve the quality of paint and varnish compositions based on aqueous film-forming dispersions in order to obtain coatings with a wider range of properties for various fields of use. One of the most effective and technically simple ways to improve the properties of paint and varnish compositions is the use of surface-active substances (surfactants), fine regulators of boundary interactions and the properties of finished materials.

In recent years, polymer surfactants have been developed and are used, which, due to their high affinity for the polymer medium, can be even more effective than their low molecular weight analogues.

Thus, the dispersibility of pigments is a total indicator characterizing the completeness, speed and energy consumption for the disaggregation of particles in a dispersion medium with their subsequent stabilization (aggregative stability). For a separate paint composition, the contribution of these processes to the resulting dispersion effect depends on the composition and structure of the film-forming agents, pigments, and modifying additives. The introduction of surface-active additives into paint and varnish compositions opens up additional possibilities for targeted changes in the physicochemical properties of the structural layers formed at the “pigment-surfactant”, “pigment-dispersion medium” boundaries and, as a result, the prerequisites for controlling the process of disaggregation and stabilization of dispersions in paintwork materials. In accordance with the classification of P.A. Rebinder, dispersing surface additives include substances that are adsorbed at the interface of two immiscible liquids or on solid interfaces, but do not form structures either in the solution volume or in the surface layers. The mechanism of the disaggregating action of surfactants in an organic dispersion medium is based on the processes of their adsorption on the hydrophilic surface of pigments.

Experimental

Surface-active effect of surfactants was established with usage of a computer-optical analyzer [8], which allows in automatic mode to determine the fractional composition of suspensions, as well as the geometric dimensions and configuration characteristics of individual dispersions. To quantify the degree of disaggregation, a calculated index d was used that characterizes the average particle diameter of pigments in suspensions.

For an adequate transfer of the image observed in the eyepiece of the microscope, an electronic converter-attachment with a magnification of × 35, equipped with a standard USB port and a software package, was used. The principle of the electronic video eyepiece is analogous to the principle of the photocell function and consists in converting light energy into electrical energy. The system unit of the attachment with small-format CCD cameras transforms the images fixed in the eyepiece of the microscope into signals acceptable for perception by the Windows XP system in personal computers.

Electronic configuration attachments are compatible with traditional microscopes. In the system developed by us, the CARLZEISS 451422 microscope was used.

At the first stage, within the software package "Spectrum of differential distribution", using the method of continuous scanning, individual image elements are recognized, then transferred to the system unit of the computer and stored (documenting).

At the second stage, in the automatic mode, the processing of the video image is performed, obtaining quantitative information about the specific number of particles (per unit area), their geometric parameters (linear dimensions, configuration, area) and, finally, in the general data on fractional composition.

The algorithm for data processing includes the following basic operations:

1. Binarization of the previously saved image –conversion of the image into black and white. In terms of Photoshop, this concept is called "by level 50%", as this selects a threshold, all values below which turn into a background color (white), and above – to the main color (black);

2 Recognition for continuous scanning and sorting of individual dispersions by the number (N, units), size and fractional composition (P, %) by reading their area in pixels (S, px). Transformation of the calculated particle size index expressed in pixels into metric units (microns).

3. Calculation of the integral and differential characteristics of the particle distribution (by their number, linear parameters and area) and their reflection in the form of diagrams, distribution functions or in tabular form depending on the optimization parameters (in the studies).

4. Derivation of functional dependences (in the form of equations or graphs) of the different integral and differential characteristics of the particle distribution (by number, linear dimensions or area), depending on the above factors. The latter makes it possible to estimate the contribution of each of them to the development of particle aggregation processes in comparative regimes, namely, the theoretical dependence (the additive function obtained with the assumption of the absence of interactions between the particles), and with respect to some basic variant (for example, in the absence of surfactants) [8, 16].

In the daily practice of the production of pigmented paint-and-lacquer materials, the use of metric particle sizes, expressed in micrometers, is accepted. In connection with the variety of shapes of pigment particles, for their size, the so-called equivalent diameter of an ideal spherical particle is accepted.

The values of d (microns) were calculated from the results of the determination (for a given multiplicity of magnification x350) of the total number of particles or their individual associates (N, pieces) and the total area (Sp, pixel) they occupy on a fixed (in the eyepiece) image in accordance with the equation:

$$\mathbf{D}_{med} = \mathbf{1,129} \cdot \mathbf{k} \cdot \sqrt{\frac{\mathbf{S_p}}{\mathbf{N}}}$$

where, Sp – area of all particles, pixel

N – total number of particles

1.129 – constant

k – conversion factor to metric units:

$$k = \sqrt{\frac{S_o}{S_{po}}}$$

For a standard sample with S_0 = 4•104 μm2 at a given (in the experiments) multiplicity of the increase by x350, the area (Sp_0) in DPI units is 32.400; value k= 1.235 μm/pixel1/2:

The disaggregating effect was additionally established by the content of fractions (P, %) of the class – (minus) 44.34 microns.

The procedure for the preparation of suspensions with different surfactant contents (0–8 g/dm^3 (for pigment weight) consisted in preliminary dissolution of a certain mass in a solvent. The resulting solutions (hereinafter A) were sent to prepare suspensions, which was carried out at a temperature of 20^{o}C in a sealed reactor (volume – 0.2 dm^3, cover factor – 0.60), equipped with an agitator (impeller agitator, frequency – 300 min-1). The quantitative contents of the pigment of titanium dioxide (1% by weight of the suspension) in the system were varied due to the change in mass loading into solutions A. To stabilize the deformation processes, the samples of suspensions analyzed with a pneumodosimeter (drop volume – 0.02 ml) were placed on a specimen slide, then fixed with a cover slip and kept under static load (10 g/cm^2) and were subjected to microanalysis for 5 minutes.

Results and discussion

During the experiments, a modified plan-matrix of the four-factor experiment was used at three levels, which was achieved by taking the factor C_{TiO2} (pigment concentration) beyond its limits. As the main factors affecting the indicators of pigment and filler disaggregation, the quantitative content of the added solvent in suspensions (C_P,%, 10÷50 volume relative to PF-060,%), surfactant ($C_{surfactant}$, in terms of the total mass of solid-phase components, - 0÷16%), the relative content of titanium dioxide with a constant mass content of pigment and filler in suspensions - 16% (C_{TiO2}, 0÷100% RH) was determined.).

The method of preparation of suspensions of paintwork materials with different content of cover-forming substance was the preliminary dilution of white spirit varnish PF-060 in the following volume ratios: 1:9, 3:7, 1:1. The resulting solutions (hereinafter-A) were directed to the preparation of suspensions, which was carried out at 20°C in a hermetic reactor (volume 0.2 dm^3), equipped with a mixing device (impeller agitator, frequency - 700 rpm). The duration of the operation, in which the stabilization of the equilibrium characteristics of suspensions and the fractional composition of the pigment and filler was fixed, was 1 hour.The relative pigment contents varied due to changes in its mass concentration in the solid phase. To estimate the disaggregating effect of surfactants, the calculated index - its concentration (in mass, %) by mass of pigment. For this purpose, a series of solutions with different surfactant content (from 0 to 16%) was pre-prepared by dissolving the surfactant in a fixed volume of solution A. The resulting solution (hereinafter - B) was sent to the preparation of suspensions. The method of preparation of modified suspensions using solution B is similar to the above.

The studied samples of paint suspensions were placed on a slide, fixed with a cover glass and maintained for a certain time under load with the help of a microdispenser (the volume of the drop was 0.02 ml). As it is known, as the solvent evaporates (in time), due to the tightening effect (in height and in plane), the development of deformation processes that affect the characteristics (geometric, structural) of the films themselves, as well as the width of the gap between the object and cover glasses, increases. The results of preliminary tests carried out on samples of different compositions showed that the load regime should be p≥10 g/cm^2. At the same time, the period of holding the samples under load was optimized, which was set according to the time required to stabilize the deformation processes in the films. For the system under study, the static load period was 5 minutes.

The response function was the total number of solid phase particles (N) falling on a fixed area of the substrate:

$$S : N = N_{relative} \cdot S,$$

The values of N in the samples were determined at a given magnification ratio (X350) using a computer-optical control system [4-6], which calculates the number of particles at S = 0.38 mm^2.

After the implementation of the active experiment the experimental array was sampled for each level of each factor within the two-dimensional matrix Yx:

$$Y = \begin{vmatrix} yx_{11}, yx_{12}, \ldots, yx_{1z} \\ yx_{11}, yx_{12}, \ldots, yx_{1z} \\ \ldots\ldots\ldots\ldots\ldots\ldots \\ yx_{m1}, yx_{v2}, \ldots, ym_{1z} \end{vmatrix},$$

m – number of levels; z - number of functional expressions for each level.

Particular dependences N = f (Ci)

$$N = f(Ci)$$

for each individual factor are shown in Fig. 1.

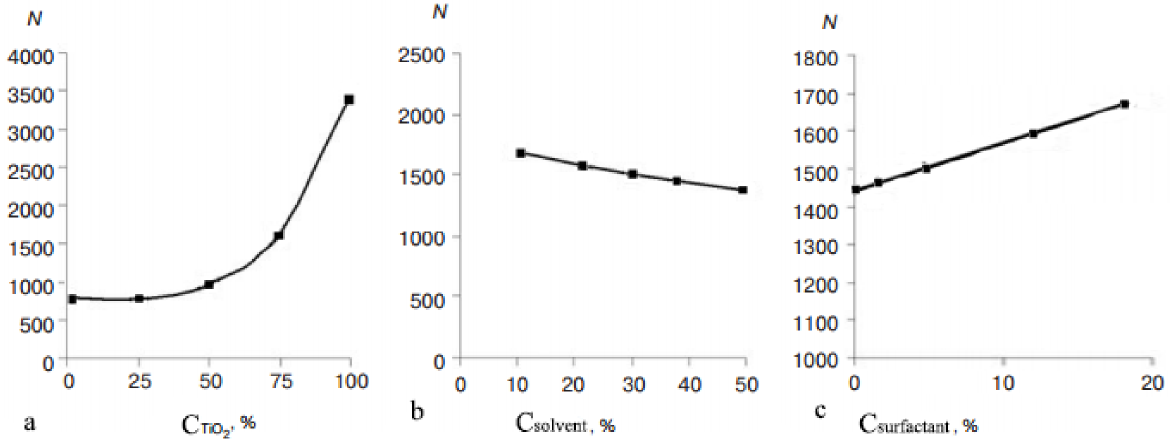

Fig. 1. Effect on pigment disaggregation rates of concentration: a - TiO_2; b - added solvent, c - AC surfactant

From the analysis of particular dependences follows a natural increase in the number of particles as the total concentration of solid-phase components in the paintwork system increases (Fig. 1, a); at the same time, the positive contribution of the as surfactant concentration to the change in N is consistent with its disaggregating effect previously established for other pigments [9-11]. Reducing the degree of disaggregation of the pigment and filler as dilution of the cover-former by solvent indicates a positive (surface-active) role of functional groups (hydroxy-, carboxy-) in the pentaphthalic resin [12-14].

After approximation of partial dependences using standard programs "Advanced Grapher" and "Microsoft Excel", one-parameter equations characterizing the effect on the response function of each factor separately (equations 1-3) are obtained.

$$N = -7{,}9 \cdot C_{solvent} + 1755{,}7 \quad (1)$$
$$N = 0{,}48 \cdot C^2_{TiO_2} - 23 \cdot C_{TiO_2} + 880 \quad (2)$$
$$N = 14{,}95 \cdot C_{surfactant} + 1436{,}6 \quad (3)$$

To build a generalized model, we used a multivariate equation of nonlinear multiple correlation, which in an implicit form has the look [10]:

$$Y_{general} = \frac{f(x_1) \cdot f(x_2) \cdot \ldots \cdot f(x_n)}{g_{average}^{\,n-1}},$$

x_1, x_2,..., x_n - factors; n - number of factors; $g_{average}$ - general average. Values of $g_{average}$were calculated by the formula:

$$g_{average} = \frac{\sum_{i=1}^{M} Y_{exp.}}{M}$$

$Y_{exp.}$ - the set of experimental data in the matrix; M - the number of rows in the matrix. After substituting the approximated expressions (1-3) into equation (4), we obtain a generalized equation that takes into account the joint contribution of all factors:

$$N = ((-7{,}9 \cdot Cs + 1756)\,(0{,}48 \cdot C^2_{TiO_2} - 23 \cdot C_{TiO_2} + 880)\,(14{,}95 \cdot C_{surf.} + 1436{,}6))/1519^2. \quad (6)$$

The adequacy of the obtained model (for the 95th significance level) was estimated on the basis of the correlation coefficients (R) and significance (t R), which were calculated by the equations:

$$R = \sqrt{1 - \frac{(n-2) \cdot \Sigma (y_{experimental} - y_{theor.})^2}{(n-1) \cdot \Sigma (y_{experimental} - y_{average})^2}}$$

$$t_R = \frac{R\sqrt{(n-2)}}{1 - R^2}$$

The calculations showed a satisfactory convergence of the experimental and calculated (according to equation 6) values of the response function:

$$R = 0{,}94, \; tR > 2.$$

Based on the solution of the generalized equation, the optimal expenditure of the surfactant, solvent and pigment in the suspension of paintwork materials were determined, providing the required degree of disaggregation of titanium dioxide. The joint contribution of the two factors to N is represented by nomograms (Fig. 2) obtained by equation 6.

The analysis of the obtained dependences shows that in order to stabilize the indices of pigment disaggregation at a fixed level, providing, for example, N = 1800, required to increase the surfactant consumption or reduce the relative pigment content as the paintwork composition is diluted with a solvent.

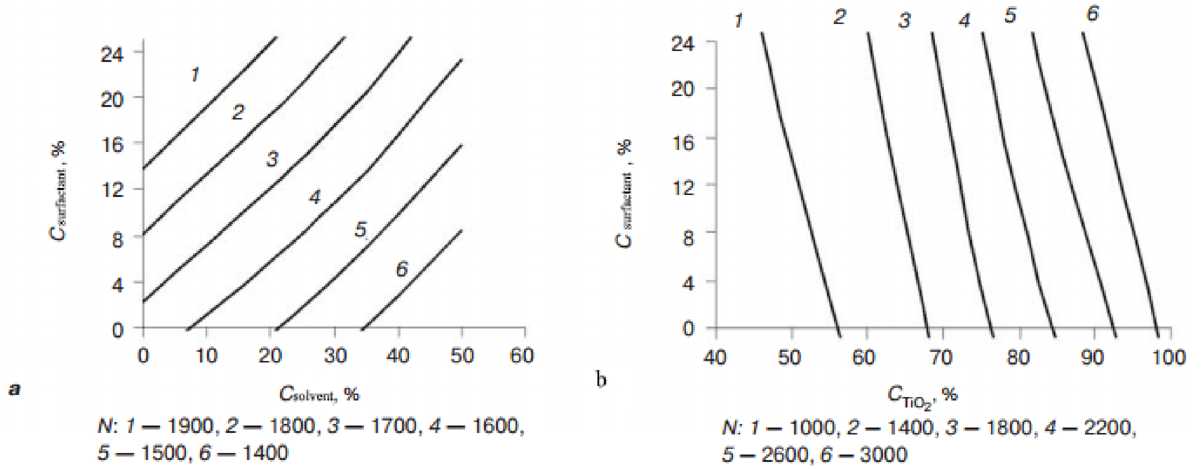

Fig. 2. Two-factor monograms: a - N = f ($C_{surfactant}$, $C_{solvent}$, C_{TiO_2} = 67,8%);
b - N = f ($C_{surfactant}$, C_{TiO_2}, $C_{solvent}$ = 30 %)

Calculations according to equation (6) showed that if Cp = 10%, then the given N = 1800 in paintwork materials is achieved at $C_{surfactant}$ = 6,95% and C_{TiO2} = 70% (relatively); a similar indicator of the degree of dispersion can be achieved by reducing the relative pigment content to 68.2%, but this will require an increase in surfactant expenditure to 12%. According to the results of balance experiments performed with paintwork compositions of these two compositions, a satisfactory convergence in the predicted and practical values of N was established; the mean-square deviation for the two examples considered does not exceed 1.2%.

Conclusions

Thus, the use of probabilistic-deterministic methods of modeling with an extended plan-matrix provides an adequate multifactorial mathematical model of the processes of disaggregation of pigments and fillers and, as a consequence, the solution of applied problems in the optimization of the composition of paintwork materials. The computer-micro-optical method applied to the analyzed in the automatic mode suspensions understudy, revealed the regularity in the development of solid-phasepigment aggregation processes and the quantitative characteristics of their distribution by fractions.

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«АҢЫЗДЫҢ АҚЫРЫ» РОМАНЫНДАҒЫ СУРЕТКЕР ЖӘНЕ ҚАҺАРМАННЫҢ РУХАНИ ӘЛЕМІ

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1960 − 80-жылдар еншісіндегі қазақ романдарына тән көркемдік сипаттарының деңгей-дәрежесін айқындай тану үшін роман жанрының әлемдік және ұлттық даму тәжірибесіне, туу, қалыптасу, өсу, өркендеу, кемелдену үлгілеріне, роман жайындағы таным-түсініктерге зер салдық, романның көркемдік-шығармашылық мүмкіндіктері мен даралық белгілерін таразыладық. Соның нәтижесінде, роман дегеніміздің өзі − өз бойына бейнелеу тәсілдері мен амалдарының қай-қайсын да қабылдай алатын, сол мүмкіндігіне орай негізгі қаһарманды сан алуан қырынан барынша жан-жақты ашып көрсетуге қабілетті, әсіресе қаһарман-кейіпкерлерді өздері өмір сүріп жатқан қоғамдық-әлеуметтік ортадағы мұраттар мен мүдделер қайшылығынан туындайтын қақтығыстар мен күрес-тартыстар үстінде ішкі жан әлемі мен сыртқы бітімін мейлінше толыққанды сипатта тұлғалай сомдауға әлеуеті молынан жететін эпикалық түр болып табылатынын