

Nail polish, in particular gel nail polish, is the most used cosmetic product. The components of the gel polish, which determine both toxicity and consumer color properties, are pigments. Zn-Al layered double hydroxides intercalated with anionic food dyes are promising pigments for use in gel polish. The parameters of the samples of Orange Yellow S-intercalated Zn-Al (Zn:Al=4:1 and Zn:Al=2:1) hydroxides synthesized at pH=8 and pH=11 were studied. The crystal structure of the samples was studied by X-ray phase analysis and thermogravimetry, and the pigment properties – by the method of measuring and calculating color characteristics in the CIELab and XYZ systems. The color characteristics of gel nail polish samples prepared using synthesized pigments were studied in a similar way.

X-ray phase analysis and thermogravimetry showed that Zn-Al-Orange Yellow S pigments synthesized at both Zn:Al ratios and pH were layered double hydroxides with the α -Zn(OH)₂ structure. The phenomenon of the decomposition of Zn-Al LDH to ZnO during the synthesis was revealed. As a result, all Zn-Al-Orange Yellow S pigment samples contained both layered double hydroxide and zinc oxide. It was shown that all samples of the Zn-Al-Orange Yellow S pigment obtained at pH 8 and 11 and the ratios Zn:Al=4:1 and Zn:Al=2:1 had high pigment characteristics and are promising for use in gel polish. The samples of gel nail polish with synthesized pigments have a red-orange color (color tone 595–604 nm) with high monochromaticity (color purity 63–75 %) and color saturation (48.7–58.3)

Keywords: Zn-Al layered double hydroxide, pigment, gel polish, intercalation, Orange Yellow S

DETERMINATION OF THE APPLICABILITY OF Zn-AL LAYERED DOUBLE HYDROXIDE, INTERCALATED BY FOOD DYE ORANGE YELLOW S, AS A COSMETIC PIGMENT

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Received date 05.09.2020

Accepted date 21.10.2020

Published date 30.10.2020

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1. Introduction

One of the cosmetic products that mankind has been using since ancient times is nail polish. Nail polish is widely used for manicure and pedicure, both for decorative and therapeutic purposes.

Decorative nail polish consists of a polymer (most often nitrocellulose [1]), solvents, plasticizers and pigments. However, at present, the most common is a gel polish, in which the curing of the nail polish occurs under the influence of ultraviolet radiation. It is the pigments that give the nail polish its color. Cosmetic pigments of gel nail polish can be both natural and synthetic. The most common mineral pigments are materials of a salt and oxide nature, for example, cobalt-chromium oxide pigments [2] as well as spinel [3–5] and pigments with the structure of various silicates [6, 7]. Inorganic pigments are characterized by high color stability but at the same time, they have a limited range of colors. In

turn, organic pigments have a wide range of colors but are characterized by low light stability.

Materials of organic-inorganic nature combine the advantages of both organic and inorganic pigments. Among the pigments of this type, the most promising are layered double hydroxides (LDH) [8] intercalated with dye anions [9, 10].

The combination of various metal cations and anionic dyes of various colors in the composition of LDH will significantly expand the color palette of cosmetic pigments. For example, Zn-Al and Acid Yellow 17 [11], Mg-Al and o-Methylene Red [12], Ni-Fe and o-Methylene Red [13], Zn-Al and Mordant Yellow 3 [14, 15], Zn-Al and Acid Yellow 3 [16], Zn-Al and Acid Green 28 bianion [17].

It should be noted that cosmetic pigments should have minimal toxicity. Therefore, the synthesis of cosmetic pigments based on LDH intercalated with food dyes of various colors is relevant.

2. Literature review and problem statement

Polymorphism was revealed and two modifications were described for bivalent metal hydroxides (except for alkaline earth metals): β -modification (chemical formula $\text{Me}(\text{OH})_2$, brucite structure) and α -modification (chemical formula $3\text{Me}(\text{OH})_2 \cdot 2\text{H}_2\text{O}$, hydrotalcite structure). For nickel hydroxide, the structures intermediate between the α - and β -forms were described in [18]. The paper [19] showed the formation of nickel hydroxide with a mixed layered ($\alpha+\beta$)-structure.

Layered double hydroxide (LDH) is an α -modification of the host metal hydroxide, in the crystal lattice of which a part of the host metal cations is replaced by the guest metal cations: for example, Zn^{2+} (the host) is replaced by Al^{3+} (the guest). Because of this, an excess positive charge is formed in the crystal lattice, which can be compensated by the intercalation of additional anions into the interlayer space. Anions of precursor salts can act as such anions. But most often, anions with special functional properties are purposefully intercalated into LDH structures. Stabilizing [20] or activating anions [21, 22] can be added to the composition of LDH. It is promising to use LDH as a nanocontainer for special anions, for example, drugs [23, 24], dye anions [25], anions for sensors [26], corrosion inhibitors [27], biologically active additives [28], food additives [29, 30] and others.

In this way, the structure of LDH includes the following main components [31]: cations of the host metal, cations of the guest metal and intercalated anions. With a purposeful selection of all three components, it becomes possible to design an LDH with the required characteristics [27].

The development of a pigment based on dye-intercalated LDH consists of three stages:

- 1) selection of the type of LDH, including the host metal cation and the guest metal cation;
- 2) selection of the intercalated dye anion;
- 3) selection of the method and conditions of synthesis.

For the synthesis of a pigment based on LDH, the most promising is the use of Mg^{2+} and Zn^{2+} as the host metal cation. Zn and Mg hydroxides are white, therefore Mg or Zn LDH represent a good basis for the clear development of the color of the intercalated dye. LDH as a pigment base must be "health-friendly" for use in cosmetics. A review paper [32] shows the minimal toxicity of Zn-Al and Mg-Al LDH.

Al^{3+} is most often used as the guest metal cation. This choice is based on high structure-forming and stabilizing properties concerning the α -modification of LDH.

The choice of a dye for producing LDH pigments of the organic-inorganic type is based on the nature of the substance. To form a pigment of the required color, a dye of the corresponding color is selected. However, for intercalation into the interlayer space of LDH, the dye must have an anionic nature. For the formation of LDH pigments, various anionic dyes are used, which significantly expands the color gamut. For use in cosmetics, the most promising are food anionic dyes: Acid Yellow 17 [11], Mordant Yellow 3 [15], Acid Green 28 [17]. Among food dyes, Orange Yellow S (Yellow Sunset) is also promising, which has a bright orange color.

In the literature, pigments obtained by intercalation are sometimes called nanocomposites. It should be noted that composite materials (organic-organic [33] or inorganic-inorganic [34]) contain a matrix and a filler with an interface between them. However, pigments intercalated with LDH

are mainly monophasic substances; therefore, it is incorrect to call them nanocomposites.

LDH was also used to purify wastewater from anionic dyes by ion exchange to obtain dye-intercalated LDH. In particular, Evans Blue was extracted using Zn-Al LDH [35], and Orange dyes – using Zn-Al LDH [36]. Various azo dyes [37], including Remazol Brilliant Violet [38], were extracted using Ni-Al LDH, blue dyes (including Maya Blue) [39] – using Zn-Al LDH, Methyl Orange [40] – using Zn-Al LDH. Amarant [41] was removed from wastewater using Mg-Al LDH, Acid Yellow [42] – using Mg-Al LDH, Congo Red [43] – using Ni-Al LDH, and Indigocarmine [44] – using Mg-Fe LDH.

The method and conditions of synthesis directly determine the micro- and macrostructure of LDH particles. Hydroxides can be obtained by chemical precipitation by the methods of direct synthesis (adding an alkaline solution to a solution of a metal salt) [45, 46], reverse synthesis (adding a solution of a metal salt to an alkali solution) [47, 48], and a sol-gel method [49]. Also, two-stage high-temperature synthesis [50, 51] and homogeneous precipitation [52] can be used for the synthesis. To obtain hydroxides, electrochemical methods are used: cathodic template synthesis [53] and synthesis in a slit diaphragm electrolyzer [54]. However, not all of the listed methods can be used to synthesize the dye-intercalated LDH pigment. To obtain such materials, two types of methods can be distinguished. The first is the two-stage type. It includes:

- 1) synthesis of LDH with inorganic anions (mainly anions of precursor salts) by the methods of reverse precipitation [11, 16, 17], precipitation at constant pH [9, 41], homogeneous precipitation [37], etc.;

- 2) intercalation of the dye anion by ion exchange [16, 17].

One of the variants of the two-stage type is the method for the reduction of LDH from LDO (layered double oxide obtained by LDH calcining) in the presence of a dye anion [12]. The disadvantages of the two-stage methods are the duration of the ion exchange as well as the need to synthesize LDH with such anions that can be easily exchanged for dye anions. It should be pointed out that the two-stage methods make it possible to synthesize pure dye-intercalated LDH. However, such methods are technologically poorly applicable in production due to the complexity, high duration and high cost.

The second variant is a one-stage type of preparation. In this case, the formation of dye-intercalated LDH occurs directly during the synthesis. For this purpose, the method of coprecipitation by reverse synthesis or synthesis at constant pH is used [55, 56]. The direct synthesis is the most promising. However, the characteristics (especially color) of pigments obtained by this synthesis have not been sufficiently studied yet.

Thereby, Zn-Al LDH intercalated with Orange Yellow S is promising for use as a gel nail polish pigment. However, the introduction of these pigments is limited by the poor knowledge of the dependence of characteristics, including color, on the conditions of one-stage synthesis. It should be noted that the characteristics of gel nail polish samples with this pigment have not been studied at all.

3. The aim and objectives of the study

The aim of the work is to determine the possibility of using Zn-Al layered double hydroxide intercalated with Orange Yellow S, synthesized under various conditions, as an effective pigment for nail polish.

To achieve the aim, the following objectives were set:

- to obtain samples of Orange Yellow S-intercalated Zn-Al layered double hydroxide under various conditions by chemical synthesis at constant pH;
- to study the structural properties of the obtained LDH samples;
- to carry out a comparative analysis of the pigment characteristics of the samples of Orange Yellow S-intercalated Zn-Al LDH to assess the effectiveness of using gel nail polish as a pigment.

4. Materials and methods for obtaining samples of Zn-Al-Orange Yellow S LDH pigments and studying their characteristics

4.1. Method for obtaining pigment samples

For the study, analytical grade qualification substances were used, except for NaOH, which was used as a granular alkali of high analytical grade qualification. As a dye, Orange Yellow S (E112, Sunset Yellow FCF) – 6-hydroxy-5-(4e-sulfonatophenylazo)-2-naphthalene-disodium sulfonate ($C_{16}H_{10}N_2Na_2O_7S_2$) was used as an analytical grade qualification substance.

For the synthesis of samples of intercalated Zn-Al LDH, a chemical synthesis method at constant pH was used, described in [55]. LDH was obtained by continuous feeding of three solutions (zinc and aluminum nitrate, alkali and Orange Yellow S, 0.5 L each) at the same rate into a reaction beaker containing 100 ml of the initial solution. The synthesis pH (8 and 11) and the molar ratio of Zn:Al (2:1 and 4:1) were chosen as the preparation conditions. The marking of the samples was as follows: E-110, pH8, 2:1, where E-110 was the Orange Yellow S intercalated food dye, pH8 was the pH value, 2:1 was the molar ratio of Zn:Al. During the synthesis, the alkali solution was taken with an excess calculated to maintain the corresponding pH. The initial solution also had a pH corresponding to the pH of the synthesis. The synthesis was carried out at a temperature of 60 °C and continuous stirring. After the introduction of the solutions, the reaction mixture was kept for an hour at the same temperature and stirred to undergo the crystallization process [57]. After that, a sample of the layered hydroxide was separated from the mother liquor on a vacuum filter. The resulting hydroxide, like polymer composites [33], was a composite material. In this case, the hydroxide itself acted as a matrix former, and the mother liquor was the filler. In this case, the composite was closed-cell. Due to the described composite structure, it was very difficult to rinse the wet LDH precipitate from the soluble salts and non-intercalated dye. Oxide and hydroxide compounds (including LDH) were highly active sorbents [58] on which a dye could be sorbed [25, 56]. For effective rinsing, a two-stage method was used: drying (70 °C, 24 hours), grinding, rinsing with distilled water and re-drying under the same conditions. Before studying the characteristics, the samples were additionally ground in a mortar and sieved through a 71 µm sieve.

4.2. Method for preparing samples of decorative gel nail polish with synthesized pigment samples

To determine the possibility of using the synthesized pigments for the production of gel nail polish, the samples of the decorative gel nail polish were prepared with the addition of this pigment [55]. The applied pigment content

was 6 % (wt.). The samples were prepared by successive grinding of the pigment with fractional addition of a colorless industrial gel nail polish base. Top Coat Soak Off (Top Coat No Sticky) was chosen as the base of the gel nail polish for the pigment application. After obtaining a sample of polish, it was applied to a glass plate and tips (2–3 layers). Each layer was cured using a specialized cosmetic UV lamp.

4.3. Methods for studying the characteristics of samples of pigments and polishes

Study of the characteristics of the synthesized pigment samples.

The crystal structure of the samples was studied by X-ray phase analysis (XPA) using a DRON-3 diffractometer (Russia) (Co-K α radiation, angular range 10–90° 2 θ , scanning rate 0.1°/s).

The thermal properties, the type of crystal lattice of the samples, and water content were determined by thermogravimetry (TG) on a Shimadzu DTG-60 thermal analyzer in an air atmosphere with a heating rate of 5 °C/min.

The maximum particle size was determined by the following method. The pigment powder was gently dispersed in vaseline oil, after which a drop of the resulting suspension was placed in a digital mechanical micrometer. When measured, the pigment particles were lined up in a monolayer, and as a result, the diameter of the largest particle was measured. The maximum particle diameter of the finished nail polish samples was measured in the same way.

The color characteristics of Orange Yellow S-intercalated Zn-Al LDH samples were studied using a spectrophotometer. For this, the LDH sample was placed in a uniform thin layer in a polyethylene ZIP bag, which was placed on an even solid base. As a result of the measurement, color characteristics were obtained in the coordinates of the international system CIE 1976 L*a*b*. Lightness (*L*) and color saturation (*S*) were also calculated in the same way. Color tone (dominant wavelength – λ) and color purity (*P*, %) were determined by the Color chart in *X, Y* coordinates.

Study of the characteristics of the nail polish samples with synthesized pigments.

The quality of the gel nail polish was determined visually by the uniformity of application and transparency of a 2-3-layer coating. The color characteristics of the gel nail polish samples with synthesized pigments were studied similarly to the pigment samples. For the study, we used nail polish samples applied to a flat glass plate.

5. Results of studying the characteristics of samples of Zn-Al-Orange Yellow S layered double hydroxide pigments

Results of studying the structural characteristics of the samples by X-ray phase analysis.

The results of X-ray phase analysis are shown in Fig. 1.

The diffraction patterns of all samples (Fig. 1, a–f) show the peaks of the Zn(OH) $_2$ lattice corresponding to Zn-Al LDH. At the same time, it has been revealed that the samples synthesized at pH=8 have lower crystallinity than the samples synthesized at pH=11. All samples, according to the diffraction patterns, contain both Zn-Al LDH (with a Zn(OH) $_2$ lattice) and ZnO (peak at 2 θ =41°). At the same time, the diffractogram of sample E-110, pH8, 4:1 (Fig. 1, c) contains three distinct peaks of ZnO.

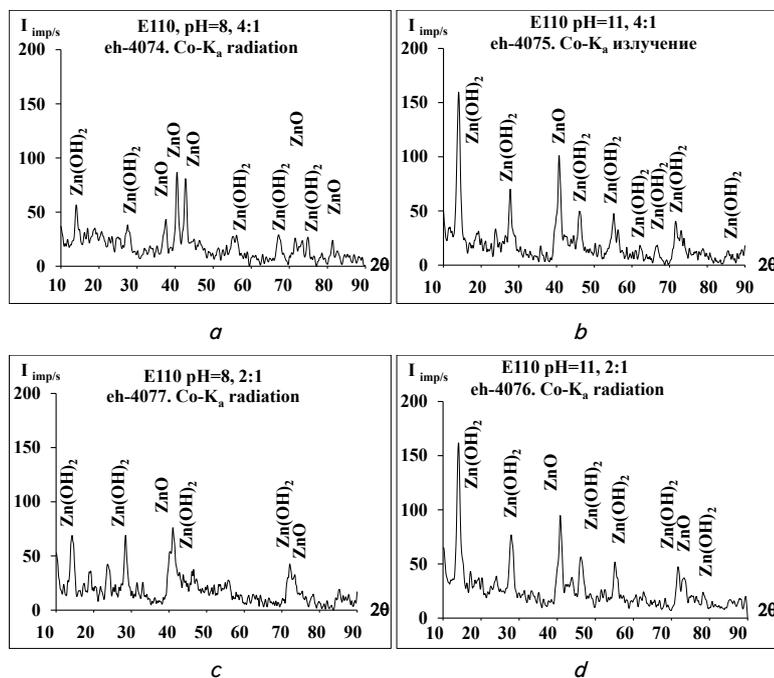


Fig. 1. Diffraction patterns of pigment samples: *a* – E-110, pH8, 4:1; *b* – E-110, pH11, 4:1; *c* – E-110, pH8, 2:1; *d* – E-110, pH11, 2:1

Results of studying the structural characteristics of the samples by thermogravimetric analysis.

All samples have similar thermogravimetric characteristics (Fig. 2). There are no clear jumps in the mass loss on the TGA curve. The total weight loss for samples synthesized at a ratio of Zn:Al=4:1 is 28.6 % and 30.9 % (for E-110, pH8, 4:1 and E-110, pH11, 4:1, respectively). The total weight loss for the samples synthesized at a ratio of Zn:Al=2:1 is slightly lower and amounts to 24.8 % and 26.7 % (for E-110, pH8, 2:1 and E-110, pH11, 2:1, respectively). For all samples, three endothermic peaks are observed on the TGA curve. The samples synthesized at the ratio Zn:Al=4:1 (E-110, pH8, 4:1 – Fig. 2, *c*, and E-110, pH11, 4:1 – Fig. 2, *d*) are characterized by an increased intensity of the second endothermic peak at a temperature of 189 °C, the first and third peaks are poorly expressed. At the same time, for the samples synthesized at the ratio Zn:Al=2:1 (E-110, pH8, 4:1 – Fig. 2, *c*, and E-110, pH11, 4:1 – Fig. 2, *d*), the intensity of all three endothermic peaks on the TGA curve is comparable. We have also observed a very intense exothermic effect on the TGA curve for all the samples. It is manifested at the temperature starting from 400–420 °C (for samples E-110, pH8, 4:1 and E-110, pH11, 4:1) and 480–550 °C (for samples E-110, pH8, 4:1 and E-110, pH11, 4:1).

Results of measuring the particle size of the pigment samples.

The measurement results are shown in Fig. 3. It is revealed that the samples synthesized at the ratio Zn:Al=4:1 (E-110, pH8, 4:1 and E-110, pH11, 4:1) have a particle size of 53–74 μm. Pigment samples synthesized at the ratio Zn:Al=4:1 (E-110,

pH8, 2:1 and E-110, pH11, 2:1) consist of significantly larger particles (maximum diameter 98–103 μm).

Study of the color characteristics of the pigments.

Visual observation has shown that samples E-110, pH8, 4:1, E-110, pH11, 4:1 and E-110, pH11, 2:1 have an orange-red color, and sample E-110, pH8 2: 1 is yellow-orange. The color characteristics of the pigment samples are given in Table 1. Color tone (Table 1) and lightness (*L*) correspond to the visually determined color and lightness of the pigments. The lightness of all pigments is in the range of 45.7–63 %. The color saturation is maximum for sample E-110, pH8, 2: 1 – 65. Color saturation for samples E-110, pH8, 4:1, E-110, pH11, 4:1 and E-110, pH11, 2:1 is very low and is 4.9, 1.2 and 3.7, respectively. It should be noted that samples E-110, pH8, 4:1, E-110, pH11, 4:1 and E-110, pH11, 2:1 are characterized by high monochromaticity – the color purity is 50 %, 52.5 % and 35 %. At the same time, the color purity of sample E-110, pH8, 4:1 is only 1 %.

Study of the color characteristics of the gel nail polish samples.

The color characteristics of gel nail polish samples obtained using synthesized pigments are shown in Table 2.

It should be noted that the color characteristics of polishes obtained with different pigments are similar. The color tone of all nail polish samples is in the range of 595–604 nm, which corresponds to the orange part of the spectrum. It should be noted that the color saturation and especially the color purity of the gel nail polish samples are higher compared to the pigments: saturation is 48.7–58.3 and color purity is 63–75 %.

Table 1

Color characteristics of samples of Zn-Al-Orange Yellow S pigments

Pigment samples	CIE Lab			Color saturation <i>S</i>	Color tone λ , nm	Color purity <i>P</i> , %
	<i>a</i>	<i>b</i>	<i>L</i> (Lightness)			
E110-, pH2:1 ,8	63.9	12	63.0	65	575	1.0
E110-, pH2:1 ,11	1.9	-3.2	45.7	3.7	630	35.0
E110-, pH4:1 ,8	-2.6	-4.2	50.9	4.9	613	50.0
E110-, pH4:1 ,11	0.3	1.2	53.0	1.2	615	52.5

Table 2

Color characteristics of gel nail polish samples with Zn-Al-Orange Yellow S pigments

Pigment sample in gel polish	CIE Lab			Color saturation <i>S</i>	Color tone λ , nm	Color purity <i>P</i> , %
	<i>a</i>	<i>b</i>	<i>L</i> (Lightness)			
E110-, pH2:1 ,8	37.4	32.4	56.2	49.5	599	70
E110-, pH2:1 ,11	38.5	29.9	55.7	48.7	604	63
E110-, pH4:1 ,8	35.0	39.0	59.6	52.4	595	65
E110-, pH4:1 ,11	34.0	47.4	65.2	58.3	596	75

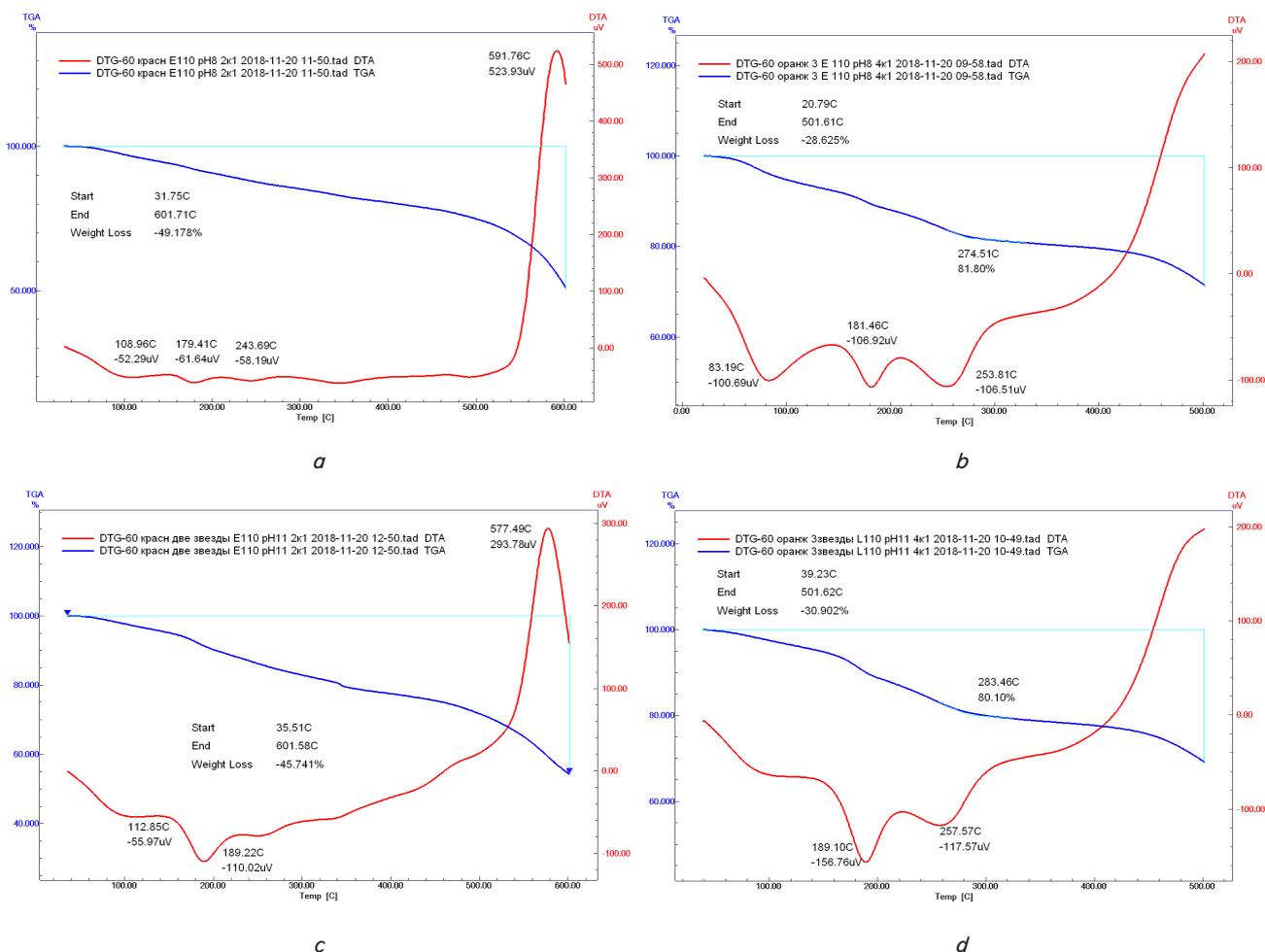


Fig. 2. TG-DTA curves of pigment samples: a – E-110, pH8, 2: 1; b – E-110, pH8, 4:1; c – E-110, pH11, 2:1; d – E-110, pH11, 4:1

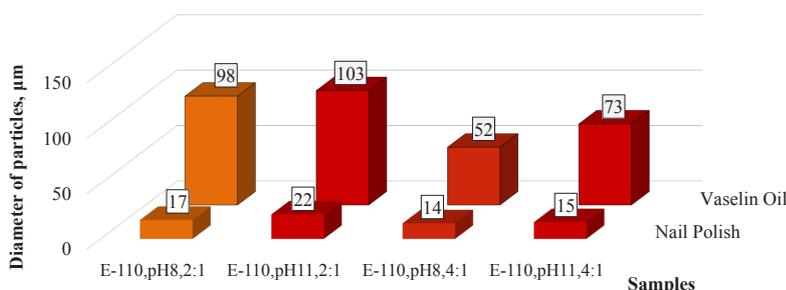


Fig. 3. Maximum particle diameter of synthesized pigments

6. Discussion of the results of studying the characteristics of Zn-Al of layered double hydroxide samples

Crystal structure of the samples. The results of X-ray phase analysis (Fig. 1) have shown that all samples of Orange Yellow S-intercalated Zn-Al layered double hydroxide have a crystal lattice of α -Zn(OH)₂. It can be concluded that all samples are layered double hydroxides of medium crystallinity. The samples synthesized at pH=11 (E-110, pH11, 4:1 and E-110, pH11, 2:1) have a large half-width at half maximum for the peak at $2\theta=14^\circ$ indicating a higher crystallinity of these samples for the plane 101. The Zn(OH)₂ crystallites in these samples might have a columnar shape. In this case, the diffraction patterns of all samples contain both Zn-Al LDH

peaks (with a Zn(OH)₂ lattice) and a peak at $2\theta=41^\circ$, indicating the presence of ZnO. Zinc oxide is likely to be formed by the partial decomposition of the hydroxide. The diffractogram of sample E-110, pH8, 2:1 contains three peaks of ZnO, which indicates the presence of a pronounced phase of zinc oxide. In the rest of the samples, zinc oxide is probably contained in the form of very thin filamentary or needle-like inclusions. The decomposition of LDH to zinc oxide, which is characteristic of samples with the ratio Zn:Al=2:1, has also been described for

tartrazine-intercalated Zn-Al LDH [55]. The results of the thermogravimetric analysis confirm the data of X-ray phase analysis. Thermogravimetric data for all samples are typical for LDH. In this case, the TG curves do not have pronounced jumps in the weight loss, which indicates either the low stability of the crystal lattice of Orange Yellow S-intercalated Zn-Al LDHs or the nanostructuring of the samples. The total weight loss (for a temperature of 500 °C) for the samples synthesized at a ratio of Zn:Al=4:1 is 30.9 % (E-110, pH11, 4:1) and 28.6 % (E-110, pH8, 4:1). In this case, the total weight loss (for a temperature of 500 °C) for the samples synthesized at the ratio Zn:Al=2:1 is less and amounts to 26.7 % (E-110, pH11, 2:1) and 23.8 % (E-110, pH8, 2:1). This indicates that the samples synthesized at pH=8 contain less crystallization water. Three endothermic peaks are observed on the TGA

curves of the samples: the 1st peak (83–112 °C) corresponds to the removal of physically bound and adsorbed water, the 2nd peak (179–189 °C) – to the removal of crystallization water, the 3rd peak (244–258 °C) characterizes the decomposition of LDH to LDO (layered double oxide). It should be noted that the crystal lattice stability of Orange Yellow S-intercalated Zn-Al LDHs is significantly lower than that of tartrazine-intercalated LDHs [55], for which the temperature of removal of crystallization water is 245 °C and decomposition of LDH to LDO – 422 °C. Samples synthesized at pH=11 (E-110, pH11, 2:1 and E-110, pH11, 4:1) have a more pronounced, high-intensity peak corresponding to the removal of water of crystallization, which indicates higher stability of the α -lattice, in comparison with the samples synthesized at pH=8. This conclusion is confirmed by the higher temperature of this peak: 189.1–189.2 °C for samples E-110, pH11, 2:1 and E-110, pH11, 4:1 and 179.4–181.5 °C for samples E-110, pH8, 2:1 and E-110, pH8, 4:1. Also, on the TGA curves of all samples, a very intense exothermic peak is observed at high temperatures, most likely associated with the combustion of the Orange Yellow S dye anion. At the same time, the temperature of the onset of the exothermic peak for samples E-110, pH11, 2:1 and E-110, pH8, 2:1 is 490 °C and 550 °C, and for samples E-110, pH11, 4:1 and E-110, pH8, 4:1 is 420 °C and 400 °C. This indicates increased thermal stability of the Orange Yellow S anion in the samples synthesized at a ratio of Zn:Al=2:1.

Pigment characteristics of the samples and nail polishes.

The measurement of the maximum size of the pigment particles has shown the differences between the samples synthesized at Zn:Al=2:1 (E-110, pH8, 2:1 and E-110, pH11, 2:1) and at Zn:Al=4:1 (E-110, pH8, 4:1 and E-110, pH11, 4:1). The former is characterized by a diameter of 98–103 μm , while the latter consists of smaller particles – 52–73 μm . It has also been revealed that samples E-110, pH8, 2:1 and E-110, pH11, 2:1 have a higher hardness and are less susceptible to grinding. This fact can be explained on the basis of X-ray phase analysis data. For the samples synthesized at Zn:Al=2:1, an increased crystallinity of the LDH component has been revealed, which increases the hardness. The softening of this increased hardness is due to the presence of zinc oxide formed as a result of the degradation of LDH during production. Probably, zinc oxide, as a soft material, plays the role of a lubricant between the solid LDH particles. As a result, when rubbed with a polymer base (top coat of gel polish), all pigment samples have been crushed to a diameter of 15–22 μm .

When discussing the color characteristics of pigments in general terms, the following should be considered. Based on the general structure of LDH, the replacement of the host cation Zn^{2+} by the guest cation Al^{3+} creates an additional positive charge in the hydroxide crystal lattice, which is compensated by the Orange Yellow S anion. Consequently, an increase in the content of the guest cation Al^{3+} (an increase molar ratio $\text{Zn}^{2+}:\text{Al}^{3+}$) should increase the content of the dye anion. As a result, color saturation should increase, and the lightness should decrease. That is, it has been initially assumed that the samples with Zn:Al=2:1 should have a more intense color. However, it has been visually revealed that all samples have a very similar red-orange color, and only sample

E-110, pH8, 2:1 has a lighter orange color. Measurement of the color characteristics of pigment samples in the CIELab and XYZ systems confirms the visual determination of the color.

The samples of gel nail polish have been made using synthesized pigments. Visual observation has shown that the prepared nail polish samples are evenly distributed pigments, the nail polish coverability is high. The measurement of the color characteristics of the pigment samples in the CIELab and XYZ systems shows that the nail polish samples have very similar parameters. It should be noted that all samples are characterized by very high color saturation (48.7–58.3) and color purity (63–75 %). This indicates a high monochromaticity of pigments, which will allow them to be used for the preparation of mixed pigments of the required color. The increase in color purity is likely due to the dispersion of the pigment particles in the nail polish base and clearer identification of the reflection spectrum of the pigment particles.

Comparative analysis allows us to conclude that all samples of Zn-Al LDH intercalated with Orange Yellow S have high pigment properties and can be used as effective cosmetic pigments for the production of gel polish. The comparison of pigments of close color gamut, Zn-Al-Tartrazine LDH [55] and synthesized Zn-Al-Orange Yellow S LDH, has shown the advantage of the pigments obtained in this work in terms of color saturation and purity as well as a wide range of production conditions. It should be noted that the application of the results of this study is limited by the fact that the synthesis of pigments has been carried out only at two pH values and two ratios of Zn:Al. In order to further improve the characteristics of the pigment based on Zn-Al-Orange Yellow S LDH, it is promising to optimize both the Zn:Al ratio and the pH of the synthesis.

7. Conclusions

1. The samples of previously not synthesized Orange Yellow S-intercalated Zn-Al layered double hydroxide have been obtained by the synthesis method at pH=8 and pH=11, and the ratios Zn:Al=2:1 and Zn:Al=4:1.

2. X-ray phase analysis and thermogravimetry have shown that Zn-Al-Orange Yellow S pigments synthesized at both Zn:Al ratios and Orange Yellow S pH are layered double hydroxides with the α -Zn(OH)₂ structure. The phenomenon of the decomposition of Zn-Al LDH to ZnO during the synthesis has been revealed. As a result, all samples of the Zn-Al-Orange Yellow S pigment contain both LDH and zinc oxide.

3. A comparative analysis of the pigment properties of the obtained hydroxides, as well as the characteristics of gel nail polish images with synthesized pigments, has been carried out. It is shown that all samples of the Zn-Al-Orange Yellow S pigment obtained at pH 8 and 11 and ratios Zn:Al=4:1 and Zn:Al=2:1 have high pigment characteristics for use in gel polish. The samples of gel nail polish with synthesized pigments have a red-orange color (color tone 595–604 nm) with high monochromaticity (color purity 63–75 %) and color saturation (48.7–58.3).

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