

У сучасному світі лак для нігтів, зокрема гель-лак, є одним з найбільш використовуваних косметичних засобів. Пігмент є компонентом гель-лаку, який визначає як токсичність, так і колірні споживчі властивості гель-лаку. Zn-Al подвійно-шарові гідроксиди, інтеркальовані харчовими барвниками аніонного типу, є перспективними пігментами для використання в гель-лаці. Вивчено характеристики зразків тартразин-інтеркальованих Zn-Al (Zn:Al=4:1 і Zn:Al=2:1) гідроксидів, отриманих синтезом при рН=8 і рН=11. Кристалічна структура зразків вивчена методом рентгенофазового аналізу і термогравіметрії, пігментні властивості – методом вимірювання і розрахунку характеристик кольору в системах CIELab і XYZ. Аналогічно були вивчені характеристики кольору зразків гель-лаку, виготовлених з використанням синтезованих пігментів.

Методами рентгенофазового аналізу і термогравіметрії показано, що Тартразин-інтеркальований Zn-Al гідроксид, синтезований при Zn:Al=4:1, є подвійно-шаровим гідроксидом зі структурою α -Zn(OH)₂. При рН=8 формується подвійно-шаровий гідроксид із низькою кристалічністю, при рН=11 кристалічність підвищується. При рН=11 виявлено розпад Zn-Al подвійно-шарового гідроксиду до ZnO під час синтезу. У результаті Zn-Al-Тартразин гідроксид (Zn:Al=4:1), синтезований при рН=11, містить як подвійно-шаровий гідроксид, так і ZnO. Для Zn-Al-Тартразин гідроксиду (Zn:Al=2:1), синтезованого при рН=11, виявлено практично повний розпад подвійно-шарового гідроксиду. Показано, що найбільш перспективними для застосування в гель-лаку є зразки Zn-Al-Тартразин гідроксиди, синтезовані при рН=8. Ці пігменти мають жовтогарячий колір (тон кольору 596–601 нм) з високої монохроматичністю (чистота кольору пігменту 60–65 %, гель-лаку – 70–75 %). Запропоновано використовувати розпад Zn-Al подвійно-шарового гідроксиду до ZnO при підвищеному рН для покращення розмельних властивостей зразків пігменту

Ключові слова: Zn-Al подвійно-шаровий гідроксид, пігмент, інтеркалювання, тартразин, гель-лак

TARTRAZINE-INTERCALATED ZN–AL LAYERED DOUBLE HYDROXIDE AS A PIGMENT FOR GEL NAIL POLISH: SYNTHESIS AND CHARACTERISATION

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1. Introduction

Decorative cosmetics had been widely used by mankind since ancient times. One of such cosmetic substances is nail polish. It is widely used for decorative and medical purposes.

Decorative nail polish consists of a polymeric base (usually nitrocellulose [1]), solvents, plasticizers, and pigments. Gel polish became the most commonly used type of polish. Its key feature is that it hardens under exposure to UV light. The color of nail polish is due to pigments in its composition. Pigments used in gel polish compositions can be of natural or synthetic origin. Among mineral pigments, the most common materials are of salt and oxide nature, such as iron oxide pigment [2], also spinel-type [3–5] and pigments of various silicate structures [6, 7]. The inorganic pigments are

characterized by high color stability but are limited by color palette. Organic pigments have a very rich color palette but are more expensive and are less.

The advantages of organic and inorganic pigments can be combined with the synthesis of organic-inorganic materials. The most promising materials of such type are layered double hydroxides (LDH) intercalated with various anions [8], including anionic dyes [9, 10].

The use of LDHs with various cation compositions with different organic anionic dyes would allow broadening the color pallet of pigments, especially for cosmetic applications. For instance, Zn-Al and Acid Yellow 17 [11], Mg-Al and o-Methyl Red [12], Ni-Fe and o-Methyl Red [13], Zn-Al and Mordant Yellow 3 [14, 15], Zn-Al and Acid Yellow 3 [16], Zn-Al and bi-anion Acid Green 28 [17].

Pigments to be used for cosmetic purposes must have minimal toxicity. Therefore, the synthesis of pigments based on food dye-intercalated LDH is of relevance.

2. Literature review and problem statement

Polymorphism is characteristic of divalent metal hydroxides with two polymorphs being described. β -form (chemical formula $\text{Me}(\text{OH})_2$, brucite structure) and α -form (chemical formula $3\text{Me}(\text{OH})_2 \cdot 2\text{H}_2\text{O}$, hydrotalcite structure). However, for hydroxides, $\text{Ni}(\text{OH})_2$ for instance, intermediate forms between α and β are also described [18]. The paper [19] described the formation of nickel hydroxide with layered ($\alpha+\beta$) structure.

Layered double hydroxide (LDH) is an α -form of “host” metal hydroxide with part of cations in the crystal lattice substituted with “guest” metal cations. For instance, the Zn^{2+} cation is substituted with Al^{3+} . This leads to the formation of an excess positive charge in the crystal lattice, which can be compensated by the inclusion of additional anions. A general formula for divalent metal-based LDH is $\text{Me}_1\text{Me}_2^{1-x}(\text{OH})_2 \cdot 0.66\text{H}_2\text{O}$, where $\text{Me}_1 = \text{Ca}^{2+}, \text{Mg}^{2+}, \text{Ni}^{2+}, \text{Zn}^{2+}$, etc., $\text{Me}_2 = \text{Al}^{3+}, \text{Fe}^{3+}, \text{Cr}^{3+}, \text{Ti}^{4+}$, etc., A – various purpose anions. Under normal conditions, these can be anions of precursor salts. But more commonly intercalated anions have special functional properties. For instance, stabilizing [20] or activating [21, 22] anions can be introduced into the LDH structure. A promising application for LDH is as nano-containers for special anions, such as drugs [23, 24], dyes [25], sensor [26], corrosion inhibitors [27], dietary supplements [28], food additives [29, 30], etc.

In practical terms, LDH is an ionic constructors with three main components [31]: “host” metal cation, “guest” metal cation, and intercalated anion. Directed choice of these components allows for the construction of LDH with required properties [27].

Development of dye-intercalated LDH pigment includes three stages:

- 1) to choose LDH type, including “host” and “guest” metal cations;
- 2) to choose anionic dye for intercalation;
- 3) to choose the synthesis method.

When preparing LDH for pigment applications, the most promising “host” metal cations are $\text{Mg}^{2+}, \text{Zn}^{2+}$. Additionally, Zn and Mg-based LDHs are white. This is a key factor in choosing “host” metal cation, as both Mg^{2+} or Zn^{2+} LDH present a good base for dye color to manifest. For pigments to be used in cosmetics, LDH, as a pigment base, should be safe for human health. In the review paper [32], it is described that Zn-Al and Mg-Al LDH have minimum toxicity.

As a “guest” metal cation, Al^{3+} is most commonly used, because of its ability to stabilize α -form of hydroxides, which LDH has.

The choice of dye for the synthesis of LDH-based organic-inorganic pigments is based on the nature of the compound. To obtain the pigment of the desired color, a dye with the corresponding color should be chosen. However, for the dye to be intercalated into LDH, the dye must be anionic in nature. Various anionic dyes are used in the preparation of LDH-based pigments, which broadens the color palette. For cosmetic applications, the most promising are food anionic dyes: Acid Yellow 17 [11], Mordant Yellow 3 [15], Acid Green 28 [17]. Among food dyes, Tartrazine is the most promising for intercalation into LDH, as it provides saturated yellow-orange color and can be combined with other dyes.

Some authors call the obtained pigments nanocomposites. However, composite materials contain a matrix and a filler, for instance, organic-organic [33] and inorganic-inorganic [34] materials, and there is always a boundary between a filler and a matrix (or binder). The intercalated LDH pigments are mainly monophase compounds, so calling them nanocomposites is incorrect.

LDH has been used to remove dyes from wastewater, thus forming dye-intercalated LDH. In particular, it is proposed to extract Evans Blue using Zn-Al LDH [35], Orange-type dyes – using Zn-Al LDH [36]. Various azo-dyes [37], including Remazol Brilliant Violet [38], had been extracted using Ni-Al LDH, light blue dyes (including Maya Blue) [39] – using Zn-Al LDH, Methyl Orange [40] – using Zn-Al LDH, Amaranth [41] using Mg-Al LDH, Acid Yellow 42 [42] – using Mg-Al LDH, Congo Red [43] – using Ni-Al LDH, and Indigo carmine [44] – using Mg-Fe LDH.

The synthesis method and conditions determine the micro- and macrostructure of LDH particles. Hydroxide preparation can be achieved by means of chemical precipitation using the titration method (addition of a basic solution to nickel salt solution) [45, 46], coprecipitation at high supersaturation (addition of nickel salt solution to a basic solution) [47, 48], sol-gel [49]. Two-stage high-temperature synthesis [50], homogeneous precipitation [51, 52] are also used. Electrochemical methods, including cathodic template synthesis [53] and slit diaphragm electrolyzer synthesis [54, 55] are also used. However, not all listed methods can be used for dye-intercalated LDH pigments. For the preparation of such materials, two fundamental methods can be outlined. The first method is composed of two stages:

- 1) synthesis of LDH with inorganic anions, using coprecipitation at high supersaturation [11, 16, 17], coprecipitation at constant pH [9, 41], homogeneous coprecipitation [37], etc.;
- 2) intercalation of anionic dye by means of anion exchange [16, 17].

A variation of such a two-stage method is the reconstruction of LDH from LDO (layered double oxide prepared by calcination of LDH) in the presence of anionic dye [12]. The disadvantages of the two-stage method are implementation difficulties, duration of the anion exchange process, and necessity to prepare LDH with anions that can easily be replaced with anion dyes. The two-stage method allows for the synthesis of pure dye-intercalated LDH. However, such methods are not suited for large-scale production due to implementation difficulties, time requirements and cost.

The second method is a one-stage direct synthesis. In this case, the formation of dye-intercalated LDH takes place directly during synthesis. For this purpose, coprecipitation at high supersaturation and constant pH is used [56]. The direct synthesis is the most promising. However, it hasn't been studied in detail, nor are characteristics of pigments (especially color) prepared under different synthesis conditions.

Thus, Tartrazine intercalated Zn-Al LDH is promising as a pigment for gel nail polish. However, characteristics, especially color, of such LDH synthesized in a single stage under different conditions are not studied well. And characteristics of gel nail polish with such pigments are not studied at all.

3. The aim and objectives of the study

The aim of the work is to determine if Tartrazine-intercalated Zn-Al layered double hydroxide prepared under

different synthesis conditions can be used as an effective pigment in gel nail polish composition.

To achieve the set aim, the following objectives were formulated:

- conduct single-stage synthesis of Tartrazine-intercalated Zn-Al layered double hydroxide under different synthesis conditions;
- study structural characteristics of prepared LDH samples;
- conduct a comparative analysis of pigment characteristics of Tartrazine-intercalated Zn-Al LDH to evaluate their effectiveness as a pigment in gel nail polish compositions.

4. Materials and methods

4.1. Synthesis of pigment samples

Analytical grade reagents were used in the study except for NaOH (granulated, Chemically pure grade). Analytical grade Tartrazine (E102, Acid Yellow 23) – Trisodium 5-hydroxy-1-(4-sulfonatophenyl)-4-[(*E*)-(4-sulfonatophenyl)diazenyl]-1*H*-pyrazole-3-carboxylate was used as a dye.

Intercalated Zn-Al LDH samples were synthesized using chemical precipitation at constant pH. LDH was precipitated by feeding three solutions (nickel and aluminum nitrates, alkali, and Tartrazine 0,5 each) at the same rate into a reaction beaker containing 100 ml of the initial solution. Two types of synthesis parameters were chosen: pH (8 and 11) and molar ratio Zn:Al (2:1 and 4:1). Sample labels are according to the pattern: E-102 pH8 2:1, where E-102 – intercalated food dye Tartrazine, pH8 – pH value, 2:1 – molar ratio Zn:Al. During synthesis, the calculated excess of alkali was taken to maintain the required pH. The pH of the initial solution was set the same as the synthesis value. The synthesis was conducted at 60 °C and continuous stirring. After solutions were added, the reaction mixture was kept under the same conditions for an hour to undergo crystallization [57]. The formed precipitate was then vacuum filtered to move the mother liquor. The formed hydroxide is a composite material, similar to polymer composites [33]. In this case, the matrix is formed by the hydroxide itself with the mother liquor acting as a filler. The composite has a closed-cell structure. Because of such structure, the precipitate is difficult to wash off from soluble salts. This requires the use of a two-stage method: drying (70 °C, 24 h), grinding, washing with distilled water and additional drying under the same conditions. Before analysis, the samples were additionally ground using a mortar and sifter through a 71µm mesh.

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

To evaluate the applicability of the synthesized pigments, samples of gel nail polish were used. Pigment content was 6 % (wt.). The samples were prepared by grinding of dry pigment and gradually mixing it with a colorless commercial gel polish base. Top Coat Soak Off gel composition was chosen as a base. After the preparation of the nail polish sample, it was applied onto a glass slide and plastic nail tips (2–3 layers). After application, each layer was cured using a gel drier UV lamp.

4.3. Characterization of pigment and nail polish samples

The phase composition of the samples was studied by means of X-ray diffraction analysis (XRD) using a DRON-3 diffrac-

tometer (Russia) (Co-K α radiation, scan range 10–90° 2 θ , scan range 10–90° 2 θ , scan rate 0.1°/s).

Thermal properties, type of crystal lattice, and water content were determined by means of thermogravimetric analysis (TG). TG analysis was conducted using a Shimadzu DTG-60 thermal analyzer under the air atmosphere and heating rate of 5 °C/min.

The maximum particle size was measured as follows. Pigment powder was softly dispersed in mineral oil, and a drop of the obtained suspension was placed in a digital micrometer. During the measurements, the particles form a monolayer, thus the diameter of the biggest particle is measured. The same method was used to measure the maximum particle diameter of the nail polish samples.

Color characteristics of Tartrazine-intercalated Zn-Al LDH samples were measured using a spectrophotometer. The thin layer of the pigment sample was placed into a polyethylene ZIP-bag, which was placed onto a flat solid base. As a result, color characteristics were measured in coordinates of the CIE 1976 L*a*b* system. Lightness (*L*) and color saturation (*S*) were calculated. Color tone value (predominant wavelength – λ) and color purity (*P*, %) were determined from the color graph in *X, Y* coordinates.

Characterization of nail polish samples with synthesized pigments.

The prepared nail polish samples were studied organoleptically by the parameters of nail polish homogeneity and stability of the 2–3-layer coat. Color characteristics of nail polish samples with synthesized pigments were studied in the same way. Nail polish samples applied to glass pieces were used for analysis.

5. Analysis results of synthesized Zn-Al layered double hydroxide pigment samples

XRD analysis results.

XRD analysis results are shown in Fig. 1.

It was found that the samples synthesized at pH=8 have lower crystallinity than those synthesized at pH=11. Sample E-102 pH8 2:1 is X-ray amorphous, and the XRD pattern of sample E-102 pH8 4:1 shows peaks of Zn(OH)₂, which corresponds of Zn-Al LDH. Sample E-102 pH11 4:1 (Fig 1, *d*) has a significantly higher crystallinity and contains both Zn-Al LDH (with Zn(OH)₂ lattice), and ZnO. At the same time, the XRD pattern of sample E-102 pH11 2:1 (Fig. 1, *c*) has no peaks of Zn(OH)₂ and only has a ZnO peak.

Results of thermogravimetric analysis.

The samples synthesized at pH=8 (Fig. 2, *a, b*) have similar thermogravimetric characteristics. Total loss is 35.23 % and 37.45 %, with no defined mass loss steps on the TGA curve. The TGA curve of sample E-102 pH8 2:1 (Fig. 2, *a*) has 3 exothermic peaks at 104 °C, 245 °C, 422 °C. Sample E-102 pH8 4:1 (Fig. 2, *b*) has a similar structure of the TGA curve and has 4 exothermic peaks at 96 °C, 169 °C, 250 °C, 367 °C. The TGA curve also has an exothermic peak at about 500 °C. Sample E-102 pH11 4:1 (Fig. 2, *c*) has a total mass loss of 24 %, and its TGA curve also does not have pronounced mass loss steps. However, the TGA curve has only two endothermic peaks at 175 °C and 235 °C. Sample E-102 pH11 2:1 (Fig. 2, *c*) shows a fundamentally different behavior: total mass loss is only 6.3 %. The TG curve has a poorly-defined mass loss step at 264 °C. The TGA curve has two exothermic peaks at 66 °C and 264 °C.

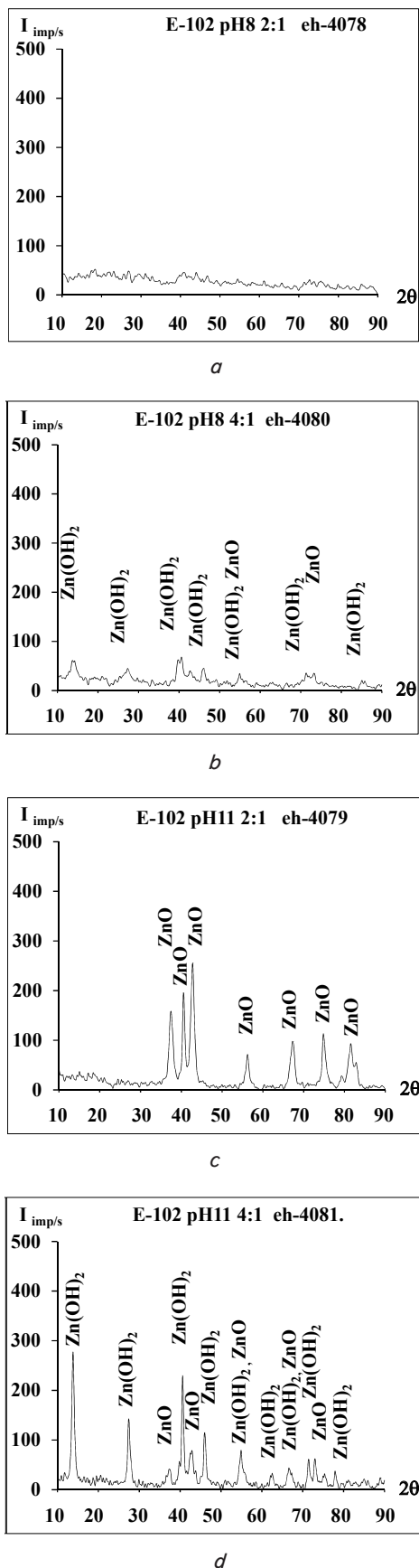


Fig. 1. XRD patterns of pigment samples: a – E-102 pH8 2:1, b – E-102 pH8 4:1, c – E-102 pH11 2:1, d – E-102 pH11 4:1

Results of particle size measurements.

The measurement results are shown in Fig. 3. It was found that the samples synthesized at pH=8 (E-102 pH8 2:1 and E-102 pH8 4:1) have a particle size of 53–74 μm. Pigment samples synthesized at pH=11 (E-102 pH11 2:1 and E-102 pH11 4:1) are composed of significantly smaller particles (maximum diameter 11–17 μm).

Analysis of color characteristics.

Visual observation revealed that the pigments synthesized at pH=8 (E-102 pH8 2:1 and E-102 pH8 4:1) have a dark-orange color, sample E-102 pH11 4:1 has a yellow color and sample E-102 pH11 2:1 is light-yellow. Color characteristics of the pigments are listed in Table 1. Color tone (Table 1) and lightness (L) correspond to the visually observed color lightness of the pigments. Sample E-102 pH11 2:1 has a lightness of 93.7%. Maximum saturation (E-102 pH8 2:1 and E-102 pH8 4:1) – 61.5. The minimum saturation value is demonstrated by sample E-102 pH11 2:1 – 14.7. It should be mentioned that samples E-102 pH8 2:1, E-102 pH8 4:1 and E-102 pH11 2:1 are characterized by high monochromaticity – color purity of 60–65%.

Color analysis of gel polish samples.

Color characteristics of gel nail polish samples obtained using the synthesized pigments are given in Table 2.

Table 1

Color characteristics of Zn-Al-Tartrazine pigments

Pigment sample	CIE Lab			Saturation S	Color tone λ, nm	Color purity P, %
	a	b	L (Lightness)			
E102- pH2:1 8	36.66	49.43	72.98	61.54	601	60
E102- pH4:1 8	30.07	53.63	76.53	61.49	596	65
E-102- pH11 2:1	1.98	14.53	93.74	14.66	585	20
E102- pH4:1 11	13.30	53.07	88.64	54.71	587.5	65

Table 2

Color characteristics of gel polish with Zn-Al-Tartrazine pigments

Pigment sample in gel polish	CIE Lab			Saturation S	Color tone λ, nm	Color purity P, %
	a	b	L (Lightness)			
E102- pH2:1 8	30.71	52.93	66.78	61.19	594	75
E102- pH4:1 8	26.05	46.61	64.30	53.40	597	70
E102- pH2:1 11	3.54	27.21	82.27	27.44	585	43
E102- pH4:1 11	18.23	60.56	73.50	63.24	587	80

Nail polish samples with pigments E-102 pH8 2:1 and E-102 pH8 4:1 have the color tone that indicates the yellow part of the spectrum. and polish samples with pigments E-102 pH8 2:1 and E-102 pH8 4:1 – color tone within the orange part of the spectrum. Saturation and color purity of nail polish prepared with the synthesized pigments are similar to those of the pigments themselves. However, their color purity is higher and reaches 75–80%.

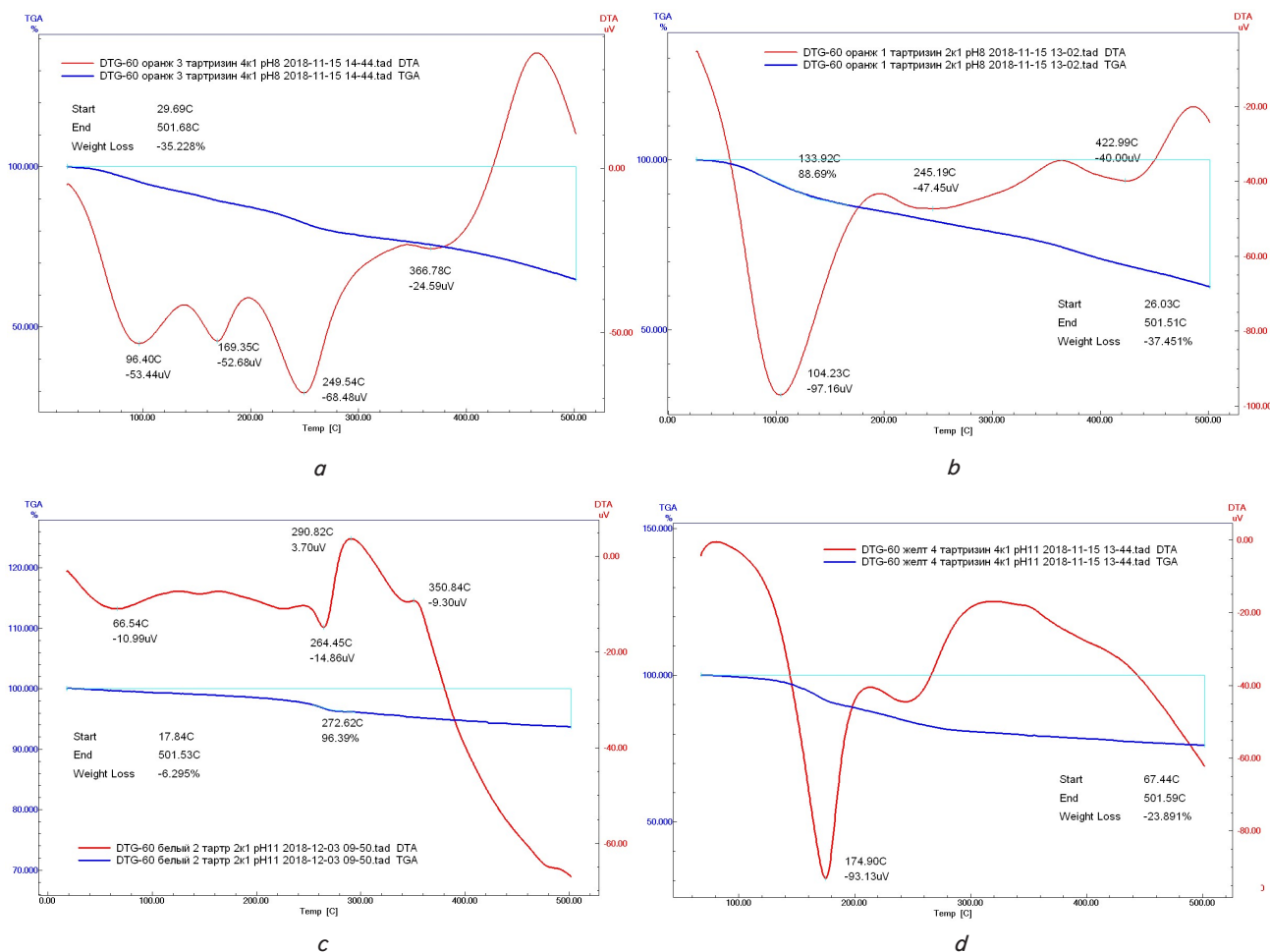


Fig. 2. TG-DTA curves of pigment samples: *a* – E-102 pH8 2:1, *b* – E-102 pH8 4:1, *c* – E-102 pH11 2:1, *d* – E-102 pH11 4:1

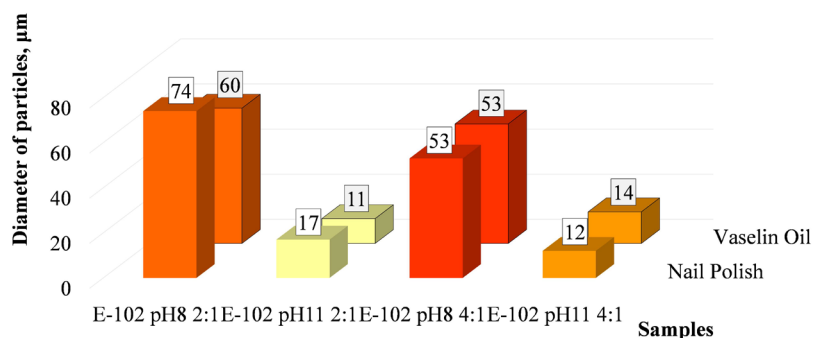


Fig. 3. Maximum particle diameter of synthesized pigments

6. Discussion of characterization results obtained for samples of Tartrazine-intercalated Zn-Al layered double hydroxide

Crystal structure. The results of XRD analysis (Fig. 1) showed that Tartrazine-intercalated Zn-Al layered double hydroxides synthesized at pH=8 (E-102 pH8 2:1 and E-102 pH8 4:1) have a crystal lattice of α -Zn(OH)₂. So it can be concluded that both samples are layered double hydroxides with low and very low crystallinity. Small half-width at half-height for sample E-102 pH11 4:1 indicates high crystallinity. The XRD pattern of this sample has peaks

that correspond to both Zn-Al LDH (with Zn(OH)₂ lattice) and ZnO. Zinc oxide is likely formed due to the partial breakdown of the hydroxide. Increasing the Zn:Al ratio from 1:4 to 1:2 (sample E-102 pH11 2:1) leads to a complete breakdown of LDH and only peaks of zinc oxide are observed on the XRD pattern (Fig. 1, c). Thus, it can be concluded that synthesis at high pH results in the breakdown of LDH, with higher pH value resulting in more destruction. Synthesis at pH=8 results in the formation of the pigment that is composed of LDH and some amount of zinc oxide. The sample synthesized at pH=11 is almost completely composed of zinc oxide with some amount of

LDH. The results of the thermogravimetric analysis support this conclusion. Thermogravimetric data for samples E-102 pH8 2:1, E-102 pH8 4:1, and E-102 pH11 4:1 are characteristic of LDH. The TGA curve of sample E-102 pH8 2:1 has 3 exothermic peaks: the 1st peak (104 °C) corresponds to physically bound and absorbed water, the 2nd peak (245 °C) – lost of crystal water, the 3rd peak (422 °C) characterized the breakdown of LDH to LDO (layered double oxide). Sample E-102 pH8 4:1 has a similar structure of the TGA curve, but it has 4 exothermic peaks at 96 °C, 169 °C, 250 °C, 367 °C. There is also an exothermic peak (500 °C) on the TGA curve, but it can also be due to the combustion of resid-

ual filtering paper. Sample E-102 pH 11 4:1 has a total weight loss of 24 %, while the TGA curve doesn't have any pronounced weight loss steps. However, it has only two endothermic peaks at 175 °C and 235 °C, which correspond to the loss of crystal water and breakdown of the LDH lattice. Peaks corresponding to the loss of physical and adsorption water are absent, which confirms the high crystallinity of the sample. Sample E-102 2:1 pH11 shows a significantly different behavior: total mass loss is 6.3 %. This indicates that the sample is primarily composed of ZnO with some insignificant amount of LDH. This is supported by a poorly defined mass loss step at 264 °C, which corresponds to the breakdown of the remaining Zn-Al LDH to Zn-Al LDO.

Pigment characteristics of samples and nail polish.

Measurement of the maximum pigment particle size revealed a significant difference between the samples synthesized at pH=8 (E-102 pH8 2:1 and E-102 pH8 4:1) and pH=11 (E-102 pH11 2:1 and E-102 pH11 4:1). The first is characterized by the particle diameter of 53–74 µm, while the second is composed of significantly smaller particles – 11–17 µm. It was also found that samples E-102 pH11 2:1 and E-102 pH11 4:1 were significantly softer and easy to grind. At the same time, the pigments synthesized at pH=8 (E-102 pH8 2:1 and E-102 pH8 4:1) had a significantly higher hardness and were difficult to grind. This fact can be explained based on the data from XRD analysis and thermogravimetry. The sample synthesized at pH=11 was found to contain a significant amount of zinc oxide, formed in the LDH breakdown during synthesis. It is possible that zinc oxide, being a soft material, played the role of a lubricant between hard LDH particles, thus easing the grinding procedure.

When discussing the color characteristics of pigments in general, one should consider the following. Based on the general LDH structure, the substitution of Zn^{2+} with Al^{3+} in the crystal lattice results in the formation of excessive charge, which is compensated by the Tartrazine ion. Therefore, an increasing content of Al^{3+} (increasing molar ratio $Zn^{2+}:Al^{3+}$) should increase the content of the dye anion leading to higher saturation and lower lightness. So, it was assumed that samples with Zn:Al=2:1 would have a more intense color. However, it was discovered that at any synthesis pH, samples Zn:Al=2:1 have a lighter tone, and samples with Zn:Al=4:1 are significantly darker. It should also be noted that samples E-102 pH11 4:1 and E-102 pH11 2:1, synthesized at pH=11, have color within the yellow part of the spectrum, while synthesis at pH=8 results into orange pigments. Measurement of the color characteristics in the CIELab and XYZ systems supports the visual observation. Based on theoretical assumptions, a decrease in color intensity indicates a lower LDH content. This conclusion is supported by the XRD results. This is especially true for sample E-102 pH8 2:1. Based on the XRD results (Fig. 1, c), the samples are primarily composed of zinc oxide. This is also supported by the thermogravimetric study – mass loss is only 6.3 % (compared to 35.2–37.5 % for the samples prepared at pH=8). These data indicate that ZnO is the primary component formed due to the LDH breakdown. As a result, Tartrazine cannot intercalate into zinc oxide. The light-yellow color of the powder implies that some amount of LDH is present. Tartrazine could also have absorbed on the

powder surface. The XRD pattern of sample E-102 pH11 4:1 also shows peaks corresponding to zinc oxide, which means that the LDH content is lower. This leads to less Tartrazine being intercalated, leading to a light-yellow color. The high color purity of samples E-102 pH8 2:1, E-102 pH8 4:1 and E-102 pH11 4:1 is also worth mentioning – 60–65 %, which indicates that the dye anion is present as a part of a single-phase, i. e. intercalated.

Gel nail polish samples were prepared using the synthesized pigments. Visual observation revealed that the prepared nail polish samples are uniformly distributed pigment, polish coverability is high. Measurement of the color characteristics in the CIELab and XYZ systems revealed that their characteristics are similar to the respective pigments. It should be noted that the color purity of nail polish with pigments E-102 pH8 2:1, E-102 pH8 4:1 and E-102 pH11 4:1 has increased to 70–80 %. This indicates a high monochromaticity of the pigments, which would enable their use in mixed pigments for the desired color. It is possible that the increase of color purity is due to particle dispersion in the polish base and improvement in the reflectance spectrum.

In summary, it can be concluded that sample E-102 pH11 2:1 cannot be used as a pigment in gel nail polish. Upon comparative analysis of color characteristics, it was discovered that the best characteristics are demonstrated by the pigments synthesized at pH=8. However, these pigments are harder. It should be noted that the application of these results is limited because the synthesis was conducted only at two pH values and two Zn:Al ratios. To further improve the characteristics of the Zn-Al-Tartrazine LDH pigment, sample synthesis at pH=8.5–9.5 and optimization of the Zn:Al ratio are promising.

7. Conclusions

1. Samples of previously not synthesized Tartrazine-intercalated Zn-Al layered double hydroxide have been synthesized at pH=8 and pH=11 and Zn:Al=2:1 and Zn:Al=4:1 ratio.

2. The results of XRD analysis and thermogravimetry revealed that Zn-Al-Tartrazine hydroxide synthesized at Zn:Al=4:1 is a layered double hydroxide with the $-Zn(OH)_2$ structure. Synthesis at pH=8 results in the formation of pure LDH with low crystallinity, and at pH=11 crystallinity increases. It was discovered that Zn-Al LDH breaks down to ZnO during synthesis at pH=11. As a result, Zn-Al-Tartrazine hydroxide (Zn:Al=4:1), synthesized at pH=11, contains both LDH and zinc oxide. For Zn-Al-Tartrazine hydroxide (Zn:Al=2:1), synthesized at pH=11, an almost complete breakdown of LDH was observed, and the samples were found to be composed of ZnO and the remaining amount of $Zn(OH)_2$.

3. Comparative analysis of the pigment properties of prepared hydroxides and characteristics of gel nail polish compositions prepared with them has been conducted. It was found that the most promising for the preparation of gel nail polish are Zn-Al-Tartrazine hydroxide synthesized at pH=8. These pigments have an orange color (color tone 596–601 nm) with high monochromaticity (pigment color purity 60–65 %, color purity of gel nail polish 70–75 %). An assumption was made that the breakdown Zn-Al LDH to ZnO discovered at higher pH can be exploited to prepare pigments with improved grindability.

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