\_ METHODS OF NATURAL SCIENCES IN THE STUDY \_ OF CULTURAL HERITAGE OBJECTS \_

# Paul Gauguin, "Tahitian Pastorals": Study of Painting Materials

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**Abstract**—A study is made of Paul Gauguin's painting "Tahitian Pastorals" painted by him in 1892–1893, during his first stay in Tahiti. The painting was carried out on canvas in the technique of oil painting. To study the stratigraphy of the layers and the pigment composition of the paint layers, the method of polarization microscopy and energy-dispersive X-ray microanalysis are used. The results of the study show that chalk from silt deposits was used as the primer. Very thin paint layers, the thickness of which varies from 10 to 30  $\mu$ m, are applied in one layer on the primer. Pigments such as viridian, ultramarine, cinnabar, arsenic copper, zinc white, lead white, chalk, and barite are revealed in the paint layers. Using the method of pyrolysis-gas chromatography-mass spectrometry, it is shown that beeswax was used as a transparent coating on the surface of the painting, which penetrated into the thickness of the paint layers and the primer of the painting during its life. The conducted research allows the presence of later layers of varnish, restoration records and residues of strengthening glue over the wax coating to be determined. The results of the study of the painting "Tahitian Pastorals" are of practical importance for further study, restoration, storage and transportation in case of the exhibition of paintings in other museums of other works by Gauguin from the collection of the State Hermitage.

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## **INTRODUCTION**

The painting by Paul Gauguin "Tahitian Pastorals" (Fig. 1) from the collection of I.A. Morozov was created in December 1892 during his first stay in Tahiti (1891–1893). The artist attached special importance to the canvas; he saw in it the beginning of a new painting period [1]. Gauguin wrote in his letter: "I have just finished three canvases, they are some of my best, and since it will be January 1 in a few days, I dated one of them, the best one, to 1893. As something rather unusual, I gave it the French name "Pastorales Tahitiennes," because I cannot find the corresponding name in the Kanak language" [2].

In the apt statement of Dzhirat-Vasyutinskii about the paintings of Gauguin we find: "These matte, delicately textured surfaces were a rejection of the dominant salon aesthetic of illusionistic oil painting and struck contemporaries with decorativeness and primitivism, reminding them of old, non-oil materials such as tempera or fresco" [3].

The painting depicts two Tahitian women, one of whom is playing a Maori reed flute called vivo, while the other is listening. In Tahitian Pastorals, the theme of Maori music was taken up, already reflected in the three works that immediately preceded this canvas. Closest to the "Tahitian Pastorals" is a painting from the Musée d'Orsay, "Arearea. A mischievous joke" 1892 [1]. According to the catalog "Tahitian Pastorals" entered the Hermitage in 1948 from the State Museum of New Western Art. Previously, the canvas was: from 1893 in the Durand-Ruel gallery; after the exhibition-sale of paintings and drawings by Gauguin at the Hotel Drouot on February 18, 1895 in a private collection in Paris; then at the Bernheim-Jeune Gallery; later in the Vollard Gallery; and since April 1908 in the collection of I.A. Morozov (purchased from Vollard for 10000 francs) [1].

For most of the paintings created by Gauguin in Tahiti, an unstable state of painting is characteristic, which could be caused by the conditions in which



Fig. 1. Painting by Paul Gauguin "Tahitian Pastorals."

these paintings were created. The extremely humid climate of the tropics, the lack of stocks of art materials, the use of a depleted binding medium in the paint layers and glue in the primer to achieve stylistic ideas to the detriment of the traditional oil-painting technique, and difficulties in delivering paintings to France. All of this to some extent influenced the further preservation of paintings. These factors were also reflected in the "Tahitian Pastorals."

In addition, earlier the painting was subjected to a series of restorations. According to information received from the Archive of the Laboratory of Scientific Restoration of Easel Oil Paintings, it is known that three restorations of the painting have been carried out over the past 45 years. Restorations in the State Hermitage were carried out within the framework of traditional conservation methods using a honey–sturgeon-glue strengthening composition, followed by the application of a varnish layer. In the past, restorations were limited only to conservation measures without preliminary research. We do not have any other information about possible restorations of the painting prior to its receipt by the Hermitage.

Given the significant interest in the work of Gauguin, which manifests itself throughout the world with an increase in the number of exhibitions, we note that in this way the painting is exposed to additional risks associated with transportation and movement. Concern for the state of preservation of Gauguin's paintings prompted us to study the cause of the unstable state of the paint layers. A lack of sufficient information about the structure of the paint layers and the materials used by Gauguin in Tahiti prompted a study of the painting techniques. Studies during the restoration of one of the paintings by Gauguin, carried out using methods such as macro X-ray fluorescence scanning, Raman spectroscopy and infrared (IR) Fourier spectroscopy with frustrated total internal reflection (FTIR-IR-Fourier spectroscopy) and without the fabrication and study of thin sections, did not allow either determination of the stratigraphy of the painting nor to understand whether the artist used mixtures of pigments and whether layers of pure pigments were superimposed one on top of the other [4]. The rest of the information regarding the technique and materials of Gauguin's painting is based not on scientific and technical research, but on information from his correspondence with friends and the dealer, revealed in [5]. Gauguin's correspondence gives a fragmentary idea of the materials and technique, since this aspect was not the most important for expressing the thoughts and feelings of the artist. Nevertheless, from the letters one can learn about the technical tasks set, his preferences in the choice of canvases, primers, paints, of course, within the framework of the possibilities of what he brought with him, was available on the islands or was delivered at his request from France [6]. The selection of material was sometimes random, e.g., local burlap was used, and the purchased paints, due to the difficult financial situation of the artist, were cheap and of poor quality.

The main purpose of the study is to investigate the materials and techniques of painting by Gauguin to understand the possible reasons for the weakening of bonds in the structure of the painting, which manifests itself in the unstable state of the painting "Tahitian Pastorals," to determine and optimize the conditions for preserving the studied work in the future.

#### **EXPERIMENTAL**

The material for research was microfragments of painting taken from by Gauguin's "Tahitian Pastorals," as well as organic material lying unevenly on the surface of the picture.

The samples were preliminarily examined under a Carl Zeiss Stemi 2000C stereomicroscope (Germany). The same stereomicroscope was used in the manufacture of microsections, which were small transparent polymer blocks, where a microfragment of painting was placed. Microsamples of fragments of the painting ( $\sim$ 30–50 µg) were used to make thin sections. The samples taken for microscopy were embedded in Tiranti transparent polyester resin (England). After completion of the polymerization process, the microsection was processed and polished using a Buehler Beta Grinder Polisher (Germany).

The study of thin section stratigraphy was carried out under a Carl Zeiss microscope. Axio Scope A1 (Germany) in polarized visible light and in the near ultraviolet (UV) region (365 nm). All photographic observations were made with a total magnification in the range of  $\times$ 50 to  $\times$ 500.

For elemental analysis, a scanning electron microscope was used. Hitachi TM3000 (Japan), equipped with a Quantax energy dispersive X-ray detector (Germany).

The composition of the organic coating film and the painting binder were studied using pyrolysis-chromatography-mass spectrometry with thermal methylation [7, 8]. For this, an Agilent 7890B chromatograph with an Agilent 5977NT MSD quadrupole massselective detector from Agilent Technologies (USA) and a PY-3030iD double-shot pyrolyzer pyrolysis system were used (Frontier Lab, Japan). The temperature of the interface of the pyrolysis installation, by means of which it is connected to the chromatograph, is 320°C.

For chromatographic separation of the pyrolysis products with simultaneous thermal methylation of the test material, an HP-5MS (5%-phenyl)-methyl-polysiloxane (30 m; 250  $\mu$ m, 0.25  $\mu$ m) capillary column was used. The analysis conditions were as follows: evaporator temperature of 320°C; split ratio of 1/20; ion source cathode switch-on time of 2 min after the start of the column-thermostat heating program. The column temperature program: initial temperature

of 40°C, maintained for 2 min, heating rate of 6°C per minute, final temperature of  $350^{\circ}$ C with holding in the isothermal mode at this temperature for 30 min. The helium-flow rate in the constant-flow mode through the column was 1.0 mL/min. The temperature at the interface between the gas chromatograph and the mass spectrometer was 320°C, the temperature of the ion source of the mass spectrometer was 230°C, and the temperature of the mass analyzer was 150°C. Electron ionization was carried out with an energy of 70 eV. The substances at the outlet of the column were recorded in the total-ion-current mode. The scanning range was from 50 to 600 amu, with a scan rate of 5 scans per second.

Small fragments of both the paint layer with primer and organic material from the surface of the painting weighing  $\sim 50-100 \ \mu g$  were transferred into a stainless steel microvessel (volume 50  $\mu$ L). Then, 7  $\mu$ L of a derivatizing agent (tetramethylammonium hydroxide in the form of a 25% solution in methanol (Sigma Aldrich)) was added to the microvessel, after which the microvessel was placed in a fume hood for 15-20 min to completely evaporate the methanol, the latter was transferred and placed in a pyrolytic installation, where helium was blown for 2 min. Then the microvessel with the sample and the derivatizing agent was lowered into the lower part of the pyrolysis installation, where the pyrolysis process proceeded. The pyrolysis temperature is 550°C. The substances formed during pyrolysis were blown into the chromatograph evaporator, from where they entered the analytical column, where chromatographic separation into individual components occurred, which were recorded at the output in the total-ion-current mode.

The recorded pyrolysis products were identified by processing the results of chromatography using the AMDIS program. Mass spectra from the NIST library were used as reference spectra.

## **RESULTS AND DISCUSSION**

In the course of the study, the composition of the inorganic components of the primer and the paint layers applied to it was studied. Figure 2 shows a thin section from the green area of the picture (at  $\times 200$  magnification) in visible reflected and UV light, as well as its electron micrograph. The study of transverse sections of the painting allows us to evaluate the stratigraphy of the paint layers. A photo of a thin section in the above modes makes it possible to more clearly see all the subtleties of applying paint layers on top of each other. In the UV photo, you can see that the overlying paint layers are deposited on a fairly thick layer of organic material.

According to the results of microscopy of a section taken from a green fragment of painting (at  $\times 200$  magnification), in visible reflected and UV light (Figs. 2a, 2b), as well as its electron micrograph and elemental

maps obtained using a scanning electron microscope (SEM) with an energy-dispersive detector (Fig. 2c, 2d), which show the presence of Ca, O, and C in the primer, it was concluded that the primer is chalk. The SEM image of the primer clearly shows the remains of rounded disc-shaped inclusions. These are the socalled coccoliths, which in terms of chemical composition are calcium carbonate. They usually have a size of several (3-8) um in diameter and are delicate calcareous plates on the surface of the cells of unicellular planktonic algae coccolithophorids; when degraded or severely damaged, they look like skeletal parts of planktonic organisms, which are hollow inside. This is typical for chalk obtained from silty marine sediments [9]. Consequently, organic chalk was used as the prime in Gauguin's painting.

Figure 3 shows the SEM spectrum of one of the fillers of a thin original green layer (its thickness reaches  $\sim 20-30 \,\mu\text{m}$ ) applied to the primer and represented by larger particles, which shows that the main elements of the green pigment are chromium and oxygen. This composition corresponds to the artificial green pigment viridian, which is hydrated with chromium oxide and which began to be produced and used in painting in the first half of 19th century [10, 11]. In addition, a finely dispersed green pigment was also found in this layer, the SEM spectrum of which is shown in Fig. 4. According to the results of elemental analysis, the inclusion contains copper, arsenic, and oxygen. According to [12, 13], such a composition can correspond to both Scheele's green (CuHAsO<sub>3</sub>), and Schweinfurt green or emerald green, which has the composition  $Cu(C_2H_3O_2)_2 \times 3Cu(AsO_2)_2$ . Both pigments have a similar elemental composition. But since by the time Gauguin created his canvases, Scheele's green had fallen into disuse due to poor pigmentation, it is likely that the artist used emerald green in this case. We note that arsenic-containing paints have harmful properties. Although at the end of the 19th century chromium oxide, which was characterized by safety and good pigmentary properties, was already produced and available, it was used less often than emerald green [14]. This is due to the fact that the cost of paints based on chromium oxide was higher than other green pigments. For example, it cost 2-3 times more than emerald green. Thus, the use of a mixture of the above two pigments made it possible to reduce their total cost, which was important for Gauguin, given his financial situation. The result obtained regarding the presence of emerald green in Gauguin's painting is confirmed by information from his letters.

On the original green layer is a thick layer of organic material, subsequently identified as beeswax, over which there is a restoration record consisting of three layers. The top layer is viridian with a small addition of Prussian blue, zinc barite and white lead. Below it is a layer with the same set of pigments, but in



**Fig. 2.** Photo of a thin section of a green fragment from the painting "Tahitian Pastorals" by Gauguin: (a) in reflected visible light, (b) UV light, (c) electron micrograph of a thin section, and (d) elemental maps of the thin section.



Fig. 3. SEM spectrum of a large inclusion in the green paint layer on top of the primer.

a different ratio, with a predominance of Prussian blue. Below is a very thin layer of viridian, under which lies a fragmentary layer of chalk. These data were obtained based on the results of elemental analysis of the indicated layers, which is shown on the elemental maps (Fig. 2d). Figure 5 shows a section from the green area of the pattern in visible reflected and UV light, as well as its electron micrograph.

According to the results of microscopy of the original yellow pigment, which is represented by crystals with a monoclinic structure in the yellow area of the



Fig. 4. SEM spectrum of a bright inclusion in the green paint layer on top of the primer.



**Fig. 5.** Photo of a section of a yellow fragment from the painting "Tahitian Pastorals" by Gauguin: (a) in reflected visible light, (b) UV light, (c) electron micrograph of a thin section, and (d) elemental maps of the thin section.

painting in visible reflected and UV light (Figs. 5a, 5b), its electron micrograph and elemental maps obtained using SEM (Figs. 5c, 5d), and also on the basis of the SEM spectrum (Fig. 6) it was concluded that to create the indicated paint layer, chromium yellow pigment (ultramarine yellow, which is lead chromate PbCrO<sub>4</sub>) [15, 16] was used. The thickness of the paint layer was ~10  $\mu$ m.

Figure 7 shows the results of microscopy of the pink pigment in visible reflected light (Fig. 7a), as well as its electron micrograph and elemental maps obtained using SEM (Figs. 7b, 7c).

On the elemental maps, at the location of the pink layer, there are elements such as Pb, Zn, Al, as well as a small amount of K. Since we do not observe a single element characteristic of red pigments, it remains to be



Fig. 6. SEM spectrum of a yellow pigment on top of the primer.

assumed that one of the red organic pigments was used, which was strongly bleached by the mixture of zinc white and white lead. Often, to create red and pink areas, artists used red organic dyes, such as madder or cochineal, planted on some inorganic sorbent [17], and the color of the pigment thus obtained depended on the composition of the sorbent. The most popular substance used for this purpose is aluminum hydroxide. The appearance of Al in the colorful pink layer indicates that  $Al(OH)_3$  was used as a sorbent.

Pigments studied in a similar way in the gray-blue area of the painting showed the presence of white lead and zinc white, barite, ultramarine and Prussian blue.

Examination of the surface of the picture showed the presence on it, in addition to the varnish, of a dense organic material with a grayish-white fluorescence in UV light (Fig. 8). The film is unevenly distributed over the surface.

The composition of the coating material on the surface was identified using pyrolysis-gas chromatog-raphy-mass spectrometry. This method is the fastest, most informative, and suitable for the analysis of organic paint materials, since it allows one to simultaneously obtain information about the presence of practically all classes of substances in the analyzed material [18].

Parallel thermal methylation allows results to be obtained with very simple pretreatment of the sample, which reduces potential losses and contamination. This procedure avoids the problems associated with the low volatility of polar compounds formed during pyrolysis. All of the above is very important, since the analyzed samples of the painting are extremely small (<1 mg) and are not rare in a single copy. In addition, very often researched pictorial objects belonging to the brush of the old masters were previously subjected to numerous restoration interventions, not always recorded, during which synthetic restoration materials could be used. This is especially true of paintings restored over the past few decades.

Figure 9 shows the chromatograms obtained as a result of studying the composition of the coating material from the surface of the painting using pyrolysis with simultaneous thermal methylation in combination with GC-MS (gas chromatography-mass spectrometry), in the total-ion-current mode (Fig. 9a); the mass chromatograms at m/z = 57, i.e., alkanes (Fig. 9b); and at m/z = 74, i.e., methyl esters of fatty acids (Fig. 9c).

The main components recorded as a result of the study of the coating film, are methyl esters of fatty acids with an even number of carbon atoms in the chain, alkanes and alkenes. Based on the library of mass spectra, palmitic, stearic, and other aliphatic fatty acids (Fig. 9c) formed upon the cleavage of ester bonds as a result of thermal methylation were identified. Palmitic acid is the most abundant  $C_{16:0}$ . Also present are long chain fatty acids with an even number of carbon atoms from 22 to 34 with a relatively high content of lignoceric (tetracosanoic) acid  $C_{24:0}$ . The presence of homologous linear long-chain alcohols  $C_{24}$ - $C_{32}$  in the form of simple methoxy ethers. In addition, a homologous series of linear long-chain hydrocarbons (Fig. 9b) with an odd number of carbon atoms in the  $C_{23}$ - $C_{33}$  chain with a maximum content of alkane C<sub>27</sub>.

According to [19–22] the substances listed above are typical of beeswax and are its markers.

In addition, the analyzed mixture contains a small amount of azelaic acid, a biomarker of oil (peak 3, Fig. 9). Unfortunately, the type of oil in this case cannot be determined, since palmitic and stearic acids, on the basis of the ratio of which the type of oil is determined, are also contained in beeswax. Rosin markers were found: dehydroabietic acid (peak 8, Fig. 9) and 7-oxodehydroabietic acid (peak 12, Fig. 9) [23]. The presence of pyrrole and 1-methylpyrrole, which are markers of glutin glue, indicates that the studied mate-



**Fig. 7.** Photo of a thin section of a pink fragment from the painting "Tahitian Pastorals" by Gauguin: (a) in reflected visible light, (b) UV light, (c) electron micrograph of a thin section, ND (d) elemental maps of the thin section.

rial contains a small amount of glue [23]. All of these listed substances, apparently, ended up in the layer under study as a result of earlier restoration measures.

Table 1 shows the recorded compounds; the number of the compound corresponds to the peak in Fig. 9.

An analysis was made of the binder of several fragments of the painting layer and the primer, from which the top wax film was previously mechanically removed. According to the results, oil, animal glue and beeswax were found in the binder.

The analysis showed that the fragility of the painting is caused primarily by the individual peculiarity of the artist's technical and technological methods. Confirmation of this is contained in a letter from Gauguin



**Fig. 8.** Photo of the surface of the painting "Tahitian Pastorals" by Gauguin with a magnification of 20 times: (a) in reflected visible light and (b) UV light.



Fig. 9. Chromatogram of the transparent material lying on the surface of the painting, in total ion current (a), mass chromatograms at m/z = 57, i.e., alkanes (b), at m/z = 74, i.e., methyl esters of fatty acids (c).

to his friend Daniel de Monfrird [2], from which it becomes clear that the problems in his paintings were laid from the moment they were created. According to information from the correspondence, the artist used rough, thick, loosely piled absorbent canvases lightly glued with animal glue. The artist squeezed out the oil binder from his paints and added turpentine to thin it. Studies have shown that chalk was used as the primer in the painting "Tahitian Pastorals." Chalk primers were popular in early European painting in the northern countries of Europe, e.g., Holland, the Netherlands, etc., and most of these paintings are well preserved, due to the fact that the chalk primer was applied to a stable wooden base, usually made of oak [24]. However, on canvas, the adhesive chalk primer does not have sufficient elasticity, it easily cracks and breaks when the picture is rolled [25]. Coarse, rough fabric fibers, thin hygroscopic primer, strongly absorbing the oil binder of paints, and very thin paint layers performed the task of creating a matte surface of the painting, but did not have stable adhesion to each other. The more moisture the chalky primer absorbs, the weaker it is bound and the more likely it is to flake off the fabric backing. Gauguin exacerbated the problem by rolling his paintings for transport, so that part of the painting was lost and required restoration even

#### KALININA, KOROBOV

The peak in Fig. 9	Substance name	Retention time, min
1	1-Methylpyrrole	8.05
2	Pyrrole	8.43
3	Azelaic acid dimethyl ester	27.93
4	Methyl ester of hexadecanoic acid	34.87
5	9-Octadecenoic acid methyl ester	37.87
6	Octadecanoic acid methyl ester	37.97
7	Tricosane	40.46
8	Dehydroabietic acid methyl ester	41.42
9	Methyltetracosyl ether	41.82
10	Pentacosane	43.19
11	Docosanoic acid methyl ester	43.59
12	7-Oxodehydroabietic acid methyl ester	44.04
13	Methylhexacosyl ether	44.43
14	Heptacosane	45.78
15	Tetracosanoic acid methyl ester	46.22
16	Methyl octacosyl ether	46.88
17	Nonacosane	48.09
18	Hexacosanoic acid methyl ester	48.47
19	Methyl triacontyl ether	49.19
20	Hentriacontane	50.36
21	Octacosanoic acid methyl ester	50.83
22	Tritriacontane	53.00
23	Triacontanoic acid methyl ester	53.66
24	Methyl dotriacontyl ether	55.21
25	Dotriacontanoic acid methyl ester	57.37
26	Tetratriacontanoic acid methyl ester	62.42

**Table 1.** Substances recorded as a result of pyrolysis with simultaneous thermal methylation of the transparent organic material lying on the surface of the picture

upon arrival in Paris [5]. Discussing the dispatch of the paintings that Monfriud was to receive, Gauguin wrote: "I'm worried about the impact of travel on the paintings and they may need to be repaired. Wash them carefully to avoid flaking paint and primer, and wax them" [6].

The presence of beeswax in the painting "Tahitian Pastorals" was confirmed by the results obtained in this work. Unfortunately, information on how wax was applied to Gauguin's paintings could not be found. Therefore, we cannot judge what caused the fact that at present beeswax is present not only on the surface of the paint layer, but also in the thickness of the primer. Perhaps the application of wax was accompanied by heat action.

A characteristic feature of beeswax is the dependence of its state on the temperature regime. Thus, at  $30-40^{\circ}$ C, it becomes plastic, and in the cold it hardens, loses its elasticity, and becomes brittle [26], thereby losing its function as a consolidating component. Thus, the presence of wax in the entire thickness of the painting must be taken into account when choosing a restoration technique and creating storage conditions.

The obtained analytical data influenced the decision to reveal the surface of the painting from later layers and made it possible to determine the difference between the artist's materials and the restoration ones.

## CONCLUSIONS

In order to obtain information about the details of Paul Gauguin's approach to creating canvases during his first stay in Tahiti, the materials and painting techniques of the painting "Tahitian Pastorals" were studied. The combined use of two methods (polarization microscopy and SEM) made it possible to establish the composition of inorganic pigments and the structure of the paint layers of the picture. The published correspondence of Gauguin attests to the early poor condition of his Polynesian paintings. The colorful layers on the test samples are very thin, applied in one layer on the primer. The soil filler is chalk of organic origin with the remains of inclusions of coccoliths. The thickness of the colorful layers varies from 10 to  $30 \,\mu\text{m}$ . The following pigments were found. White: white lead, zinc white, barite; yellow: chrome yellow; blue: ultramarine, Prussian blue; red: red organic pigment, possibly carmine, deposited onto aluminum hydroxide; and the greens are chrome green, an arsenic copper compound, possibly Schweinfurt green or emerald green.

Through the use of the gas chromatography-mass spectrometry method, the composition of the material of a thin transparent film on the surface of the painting, which was identified as beeswax, was established. The film is unevenly distributed over the surface. Due to the existence conditions, uneven penetration of beeswax into the thickness of the paint layers and primer is observed. In some parts of the picture there are records made on a layer of beeswax.

It has been established that the unstable state of the painting is connected, firstly, with the fact that an adhesive chalk primer was applied to a rough canvas, which cracks on such a basis. Secondly, the instability of the painting can be caused by the presence of beeswax in the paint layers and in the primer, which, when the temperature drops, becomes brittle and loses its consolidating properties.

The results of the study of the painting "Tahitian Pastorals" and the establishment of the reasons for the unstable state of the painting are of practical importance in the further study, restoration and storage of other works by Gauguin from the collection of the State Hermitage.

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NANOBIOTECHNOLOGY REPORTS Vol. 17 No. 5 2022

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