INVESTIGATION AND ANALYSIS OF VINTAGE HAND-COLORED PHOTOGRAPHS FROM FRANCIS AMIN'S PRIVATE COLLECTION, EGYPT

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Abstract

Prior to the introduction of color photographic process, the only method of producing photographs in color was to apply coloring agents by hand. Hand-colored photographs were very popular in the 1920s and they are still produced today. However, little research has been carried out to study the coloring agents used in terms of their chemical composition and how they respond to deterioration agents (i.e., temperature, relative humidity and UV radiation). The identification of pigments on hand-colored photographs is necessary to profoundly understand the material and technique used, and thus select proper conservation procedures. In many cases, the application of non-invasive techniques is mandatory since physical sampling of the art works is not possible. This study represents a research aimed at studying the hand-colored photographs and the identification of the pigments. For this purpose, X-ray fluorescence spectroscopy is used in detecting elements that make up the photograph and most significantly the pigments used for hand-coloring. The paper further discusses the use of multispectral imaging system as a new approach in photograph conservation field. While this technique does not provide the analytical capability that spectroscopies offer, its use has the advantage of being a rapid solution for the examination of large areas. The paper looks into another important issue of conservation which is the impact of different degradation agents on the color of the photographs. This is particularly necessary for such photographic materials since they are very sensitive to the surrounding environment. For evaluating the effect of the principal degradation agents, a group of photos was exposed to different artificial aging conditions (i.e. temperature, relative humidity and UVA). The CIELAB color coordinates (L*, a*, b*) were used to express color change.

Keywords: Hand-Colored photographic prints; Pigments; Deterioration; Color change; Artificial aging; Multispectral imaging system.

Introduction

Almost from the beginning of photography, there was a quest for color. In 1839, Louis Jacques-Mandé Daguerre announced the first practical photographic process, the daguerreotype. Immediately, the world began a love affair with daguerreotype [1]. However, a major criticism of the process was its inability to record the photographic subject in color [2]. In an attempt to correct this deficiency, it soon became practice to add color to the monochrome photographs

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while experimenters were busy seeking a way to record colors directly [3]. Photographers and colorists used dyes, pigments and metal compounds to hand color photographs [4]. The methods of hand applying color to photographic images are nearly as old as photography itself [2]. Only three years after Daguerre's announcement, the first American patent for hand-coloring daguerreotypes was granted to Benjamin R. Stevens and Lemuel Morse. Later that same year, a second American patent for coloring daguerreotypes was granted [5]. As photographic imaging processes became more sophisticated, with photographic images appearing on everything from glass to leather [6] to ceramics, so did the coloring techniques [7]. For every photographic medium, a method for hand-coloring was developed.

Photographs were colored in one of two ways: hand coloring (also known as hand tinting) and it refers to a lightly painted image that is still distinguishable as a photograph; or over-painting and it refers to an image that has been heavily painted and whose photographic origins may have been completely obscured [5, 7, 8].

Any photograph could be colored in a variety of ways, but most colorists preferred to work with photographs that were not overdeveloped and contained the entire range of tonal values [7]. Since the silver layer absorbed color differently than the rest of the photograph, the colorist would often use a coating of varnish to create an even appearance. Coloring was seen as something that should enhance the photograph and not obscure it, so it was recommended to apply a few light layers of color until the right shade was achieved rather than one dark coating [6, 7]. Most colorants needed the addition of a binding material such as gum Arabic to adhere to the photographic substrate. The colors most often used for coloring photographs were India red and pink madder for coloring cheeks, and gold paint for coloring jewellery [5].

The paper medium was most familiar to artists, and water colors were a natural colorant for paper prints [7]. Prints needed to be washed several times before painting. Albumen prints were particularly greasy and needed to be treated before pigment would adhere. The favorite method employed by photographic colorists was to lick the paper and let it dry. Most paper prints were mounted, and often the prints were varnished, before coloring to give the colors a smoother appearance. Paper prints were painted with oils, water colors or aniline colors. Aniline colors were particularly recommended for albumen prints which needed no preparation for their use due to the albumen's affinity for the dye [6, 7]. Water colors tended to leave a darker edge of color at the boundaries of the painted area. Pastels were also used to color paper prints. First the print was prepared with a thin coating of shellac, then grit was added, and finally the print was colored [9]. The adequate way to preserve the colors is through the use of glass. Charcoal or India ink was often used to retouch paper prints emphasizing details or adding in completely new material [7].

Basically, a hand-colored photographic print is composed of: a primary paper support, on which the binder layer rests; a binder (i.e., albumen, collodion, or gelatin) for holding the final image material; the final image material (i.e., silver particles and dyes or pigments); and a secondary support (i.e., boards or mats). Another component which may be included is an interlayer between the support and image layer known as the baryta layer [10, 11]. This layer is composed of gelatin and barium sulfate [12].

Photographic prints are one of the most vulnerable objects in museum, archives and libraries. Due to their complex chemical and physical nature, they are prone to deterioration and/or degradation by intrinsic and extrinsic agents [13]. Factors that affect the stability and permanence of photographic prints are inherent vice, improper temperature and relative humidity levels, atmospheric pollution, improper light levels and irradiations, biological threats, poor display and storage materials and conditions, inappropriate handling and misuse, disasters and water [8, 14, 15]. Hand-colored photographs are particularly sensitive to their environment due to the presence of pigments [16]. Hand-colored photographs are among the most light-sensitive photographs as they rapidly fade when exposed even for brief periods. Accordingly, for their long-term preservation, they require special interventions. Although exposure to all

radiation is harmful to photographs, the shorter wavelengths found in the UV portion are more damaging [17].

In the past years, very little research was dedicated to hand-colored photographs. The main aim of the present study is the identification of pigments used in several hand-colored photographic prints. Various investigation and analytical techniques were employed. Spectrophotometric analysis was carried out to evaluate the optical and chromatic characteristic of the selected photographs; while X-ray fluorescence was used to study their elemental compositions. Multispectral Imaging (MSI) was used to detect surface damage and areas of retouching, and in some cases, to characterize pigments. These techniques provide important information on the materials present and can assist with the conservation decision-making process.

Finally, different conditions of artificial aging were carried out on selected sample set to study the behaviour of hand-colored photographs in terms of how they respond to the surrounding environment [18].

Materials and Methods

Materials

Naturally aged hand-colored photographic prints were chosen as samples in order to give a more practical, rather than theoretical focus to this research work. The sample set was generously given to the authors by Francis Amin, a famous photo collector from Luxor, Egypt. The samples, reported in Table 1, were made during different time periods; and therefore they were categorized into the following four groups:

1. The first group, called the "OLD" group, includes five photographic prints dating back to the period from 1850 to 1900.
2. The second group, called the "OLD 1" group, includes five photographic prints dating back to the period from 1900 to 1950.
3. The third group, called the "NEW" group, includes ten recent photographic prints dating back to the period from 1950 – 2000.
4. The fourth group, called the "TEST" group, includes five photographic prints of non-significant historic or artistic value. This group will be exposed to destructive aging tests since it will be possible to cut smaller samples and use them for a   more controlled testing approach.

In the preliminary phase of the research, some points were selected on each sample, on which measurements of investigation and analysis were performed.

Methods
Multispectral imaging
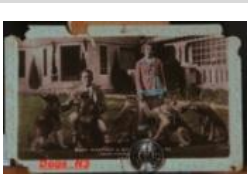
Multispectral imaging (MSI) was conducted, not only for a documentation purpose, but also to obtain data useful for better characterization of the pigments used. Multispectral camera is used to capture a range of wavelengths, each of which determines a specific feature of the analysed object. The MSI images presented in this paper were acquired with an Artist camera system of Art Innovation equipped with system CPS 200E. The technique followed in this work uses a filter wheel approach to capture images of different bands of the electromagnetic spectrum. Seven glass filters are used in the motorized filter wheel placed in front of either a 2 or 5M Pixel multispectral imaging system with a 1600×1200 pixels CCD sensor.

Images for this study were acquired in two spectral bands: visible, VIS (380–780nm); and Infrared, IR (780 – 1100 nm). The first image is a true-color image which shows the examined surface in actual colors. It covers the full visible spectrum using only Red, Green, and Blue channels, mapped to their respective colors (RGB images). The second image is a false-color image using the near Infrared part of the electromagnetic spectrum (900nm), IRFC

(Infrared False Color). The IRFC image allows differentiating between pigments with same hue but different chemical composition.

To ensure a reproducible and true-color result, the multispectral camera system is first calibrated by placing a neutral grey card Fotowand 4964 in front of the camera. The distance of the camera to the table surface can be manually adjusted to change the field-of-view and guarantee an optimally sharp image. The used light sources are two halogen lamps equipped with the instrument. They were collocated at a distance of 70cm and at 45° respect to the samples (Table 1).

Table 1. The photographs belonging to the four groups and their identification code

OLD GROUP

| Woman_01 | Napoli_02 | Woman_03 | Paris_04 | Woman_05 |

OLD 1 GROUP

| Man_01_1 | Love_01_2 | Woman_01_3 | Woman_01_4 | Woman_01_5 |

NEW GROUP

| Horse_N1 | Boy_N2 | Dogs_N3 | Woman_N4 | Child_N5 |

| Cafè_N6 | Woman_N7 | Woman_N8 | Man_N9 | Man_N10 |

TEST GROUP

| Woman_T1 | Woman_T2 | Woman_T3 | Woman_T4 | Woman_T5 |

Spectrophotometric measurements

An important issue of conservation is the impact of environmental conditions on the original color of the objects being stored or displayed. In order to analyse the absolute color changes due to exposure to different aging conditions, a Konica Minolta spectrophotometer, model CM-2006d, was used. The analysis was conducted with measurement geometry d/8°, selecting an area of 3mm in diameter (SAV, Small Average Value). The results are related to the D65 illuminant and the CIE 1964 standard colorimetric observer (10° standard observer). In the following text all colour coordinates report then subscript "10" [19].The values were obtained from repeated measurement (5 different acquisitions) and the elaboration regarded SPEX/100 data (specular component excluded and UV included). The uncertainty is calculated as Root-Mean-Square (RMS) average of Standard Deviation (SD) of repeated measurements and systematic uncertainty attributable to instrumentation. This last contribution was estimated on the basis of CIELAB colour coordinates measured on White Calibration Plate.The acquisition step was achieved using the SpectraMagic® software, while Origin® software (OriginPro 8) was used for data processing.

The PH3DRA (Physics for Dating Diagnostics Dosimetry Research and Application) laboratories routine protocol [20] was used for spectrophotometric measurement. The scale adjustment represents a very important step [21] and it was performed using the White Calibration Plate (CM-A145) as a target for the maximum lightness and the device CM-A32 for the minimum lightness. The results were elaborated with attention to the values of SRF% vs wavelength in blue (400-500 nm), green (500-600 nm) and red (600–700 nm) regions and the analysis of the color coordinates: $L*$, $a*$, $b*$, belonging to the CIELAB space. These coordinates were used to express color change, where $L*$ defines lightness and varies from 0 (black) to 100 (white). The $a*$ scale denotes the red/green values, where $+a*$ means red and $–a*$ means green. The $b*$ scale measures yellow/blue, where $+b*$ means yellow and $–b*$ means blue [22, 23].

In the cultural heritage field, the maximum acceptable difference of $\Delta E_{ab}*$ has to be such to not compromise reading of the artworks. In particular, for the quantification of the colour difference after aging tests [24, 25], the $\Delta E_{ab}*$, 10 quantity is selected. The choice is based on the assumption that it represents the most used in the cultural heritage field [26, 27]. With regard to the $\Delta E_{ab}*$ uncertainty, this was calculated by uncertainties propagation theory [28, 29].

X-ray fluorescence analysis (XRF)

In studies of fine art photography, both modern and historical, XRF is used primarily to identify the photographic processing chemistry and the nuances associated with chemical composition of the image layer. The XRF analyses have been performed utilizing the fluorescent X spectrometer, Lithos 3000 (ASSING). It is one of the smallest existing analysis systems, and it is ideally suited for in situ analysis and requires no sample preparation. The X-ray source is constituted by a conventional tube, air forced cooling type, operating at a voltage of 30kV maximum acceleration and a maximum current of 0.5mA.

The detection system is constituted by an energy dispersive detector 600 μm thick and with 7mm^2 of active area, with a resolution of 160eV at 5.9keV. The pointing system is constituted by a laser interferometer, which measures the distance between instrument and sample. This system allows the positioning of the fire point with an uncertainty of ±15μm.

For each sample, the parameters used for the analysis were: 20kV and 100 seconds of acquisitions. The information about the sample comes not only from the surface but also from the underlying layers, according to a penetration depth that depends strictly on the elemental composition of the surface and can therefore vary from point to point.

Artificial Aging

Samples were exposed to selected conditions of temperature, relative humidity and light in order to study their effect on the samples in terms of color change, since they are significant

factors affecting the permanence of photographic collections whether they are on display or in storage. Samples were exposed to the following two types of artificial aging conditions for evaluating the induced photo-degradation and the thermo-hygrometric degradation:

1. Humid heat aging was carried out to evaluate thermo- hygrometric degradation at a temperature of 80±2°C and a relative humidity of 65% for a period of 74 hours based on conditions described in specification ISO 5630-3:1996 for paper-based materials [30]. The test was performed in a Xenotest Alpha climatic chamber that allows the direct setting and control of relative humidity and of the temperature values.

2. Light aging was performed to evaluate induced photo- degradation. The Ultraviolet lamp emitted with a wavelength at about 320nm. The total hours of photo-aging are 26 hours. This test was conducted using a Verivide's Color Assessment Cabinet. It offers immediate colour viewing in consistent, controllable lighting conditions and it removes problems caused by the variability of natural daylight. This is an indispensible tool for color assessment, color quality control and comparison of color variation.

Results and Discussion

The results are presented in two main points. The first discusses the characterization of the samples and the identification of the pigments used as coloring agents. The characterization was carried out with multispectral acquisitions (RGB and IRFC images) for a documentation purpose, but also to obtain data useful for better characterization of the pigments used. The identification of the pigments was possible to cross the data obtained from spectrophotometric and XRF analysis. The second is related to the colorimetric measurements post accelerated aging tests. This last step is important to deepen aspects related to conservation purposes.

Characterization of hand-colored photographs and pigment identification
Multispectral acquisitions

As an example of the old group set, sample Woman_01 is presented as false color IR and RGB images (Fig. 1). By comparing false color Infrared images for the red pigment in photo (woman _01) to the one used in photo (paris_04), we found that although both photos have the same red tone, in false color IR they show different yellow tones, indicating that they are of different chemical composition (Fig. 2). As example of TEST group, the sample Woman T_1 is presented (Fig. 3).

Fig. 1. RGB image (right) and false-color infrared image (left) for sample Woman_01

Fig. 2. RGB image (right) and false-color infrared image (left) for sample Paris_04

Fig. 3. RGB image (right) and false-color infrared image (left) for sample Woman_T1

Spectrophotometric analysis

Spectrophotometric measurements were carried out on all samples used in this study to present the comparative surface color properties of each pigment. In particolar, the analysis regarded the trend of spectral reflectance factor (SRF%). Chromatic values for the OLD GROUP sample set are shown in Table 2. All reported values of chromatic coordinates have uncertainties of 4%.

Based on the obtained results, the blue pigments used for hand-coloring the photographs belonging to the first group are not similar. Results also showed that two types of red pigments are used. As for the yellow pigment, comparison between SRF% shows that they are of similar chemical composition. This result was later confirmed by X-ray fluorescence spectroscopy. Figure 4-5 show the trend of spectral reflectance factor (SRF%) for two photographs.

The identification of the pigments is possible through the combined results of the database SRF% trends and the information obtained by other techniques (i.e. XRF). It is possible thank to the use of the mask, a tool ensuring that the same colored points are investigated with the two techniques.

Table 2. Chromatic values for the OLD GROUP sample set

| ID code samples	$L*$	$a*$	$b*$	$C*$	$H*$
OLD GROUP					
Woman_01					
Y5	72.0	2.9	22.6	22.8	82.5
B	56.8	-9.6	-2.1	9.8	192.4
R3	48.5	31.9	12.0	34.1	20.6
R1	68.5	11.0	22.4	25.0	63.7
Y4	57.1	6.1	25.1	25.9	76.2
Paris_04					
B1	64.6	1.00	24.6	24.6	89.7
Y1	62.0	17.0	73.0	74.9	76.8
R2	31.0	38.7	18.7	43.0	25.7
B2	63.0	0.7	19.0	19.0	87.8
Napoli_02					
B1	63.6	0.3	24.4	24.4	89.1
R2	59.0	11.4	29.8	31.9	68.9
G3	61.9	4.9	31.3	31.7	80.9
W4	69.0	6.2	34.0	34.6	79.5
Y5	72.5	2.9	27.9	28.1	84.0
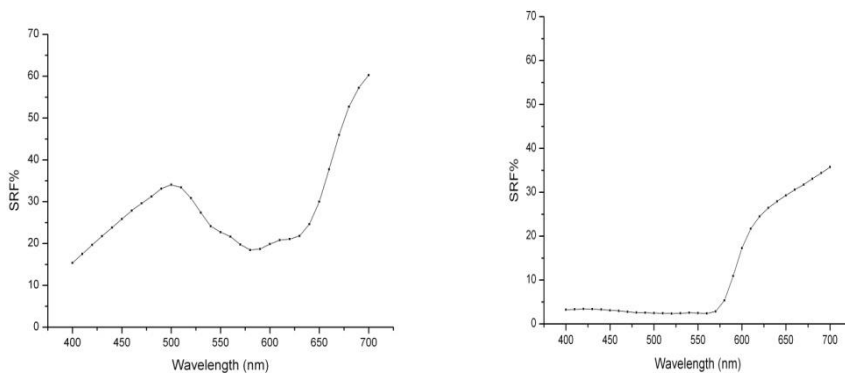
Fig. 4. SRF% trend for sample Woman_01, shown in Figure 1, blue (right) and red (left) pigments
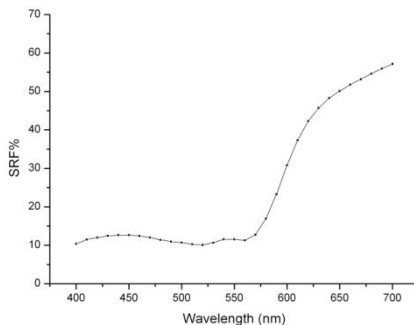
Fig. 5. SRF% trend for sample Paris_04, shown in Figure 2, (red pigment)

X-ray fluorescence spectroscopy

Two prints (i.e. Woman_01 and Paris_04, both belonging to the old group) were selected to comment the X-ray fluorescence spectroscopy results. These samples were chosen because

they are the most representative of the principal hues for the photos of all groups. The analysis was only qualitative and was aimed to identify the major elements.

Results reveal the presence of barium and calcium (Fig. 6). Both elements were found in most analyzed samples. Both are related to the composition of the paper. Barite is a special barium sulphate coating, known as the baryta layer that is traditionally applied to a fiber-base photographic paper prior to coating with the emulsion layers [31]. Although this layer is important in producing a smooth or glossy print, the baryta paper of the 1890s did not result in the luster or gloss print surface that became the standard for fine art photography in the twentieth century [32].

The spectrum for the red pigment used in sample Paris_04 reveals the presence of mercury (Hg), which is related to the cinnabar pigment (Fig. 7). The red pigment cinnabar (HgS) has been used as a painting material for thousands of years in many cultures [33]. By analyzing another red pigment found in sample Woman_01 (Fig. 8), the resulting spectrum revealed the presence of arsenic (As) indicating that the red pigment used is most likely realgar (As4S4), a red–orange used by artists from different cultures since antiquity [34].
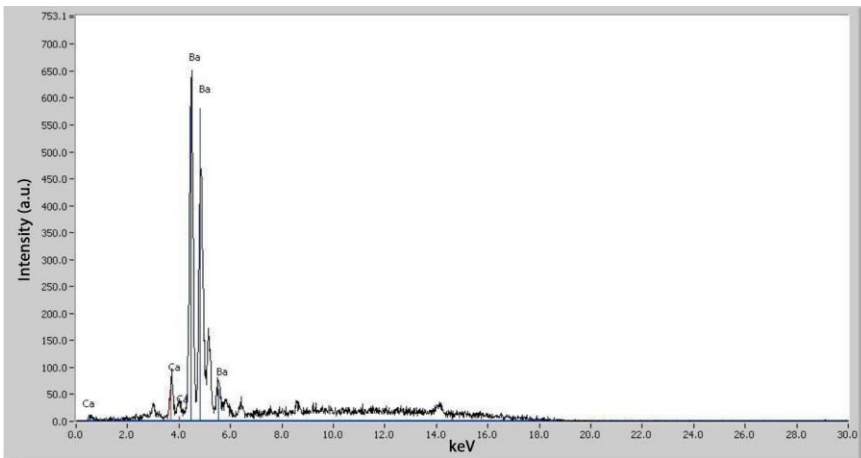

Fig. 6. XRF spectrum of a sample belonging to the OLD GROUP set
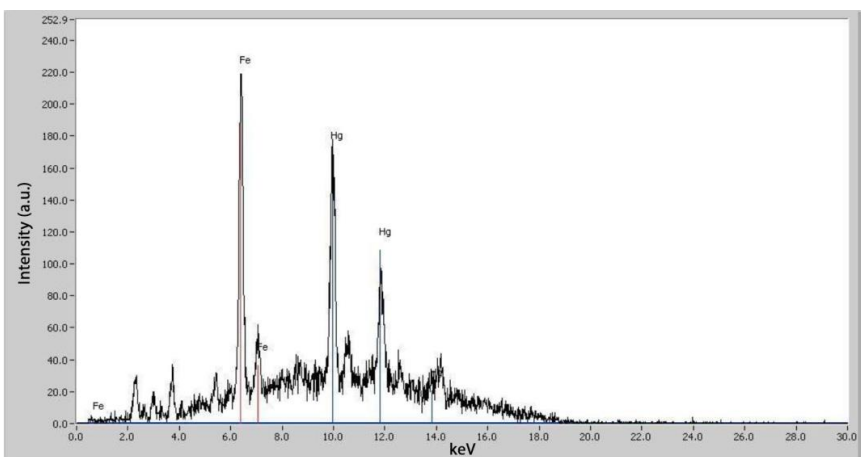

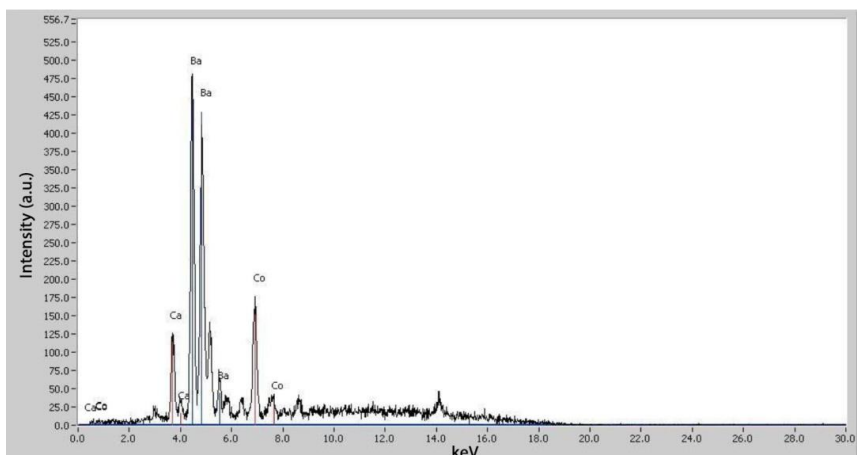**Fig. 7.** XRF spectrum of red pigment of Paris_04 (Old Group)

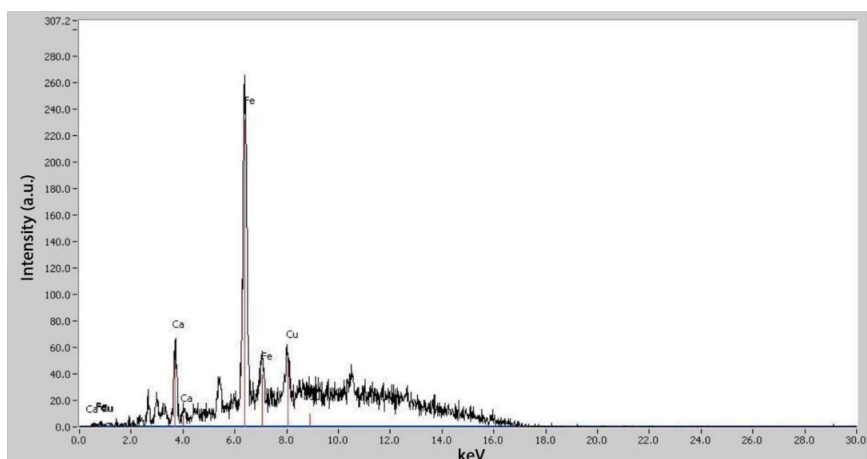**Fig. 9.** XRF spectrum of blue pigment of Woman_01 (OldGroup)

Fig. 10. XRF spectrum of yellow pigment of Paris_04 (OldGroup)

The obtained results for the blue pigment used in sample Woman_01 (Fig. 9) reveals the presence of cobalt indicating that cobalt blue was used. Cobalt blue is an inorganic oxide of cobalt and aluminum. Cobalt blue dark and cobalt blue light ($CoO \cdot Al_2O_3$) pigments were already used in ancient Egypt for decorating pottery. Cobalt blue greenish ($Cr_2O_3 \cdot CoO \cdot Al_2O_3$) was produced in the middle of the 19th century. The color of pigment varies from bluish green to greenish blue depending on the molar ratio of aluminum and chromium [35].

The identified yellow pigment in sample Paris_04 (Fig. 10) is most likely yellow ochre, this result was suggested due to the presence of iron (Fe) [36].

Evaluation of color change post aging

Among the samples belonging to the new group, sample Woman_T1 was selected, as an example for discussing the evaluation of color change post aging. This sample, in fact, has the majority of colors; and therefore is a representative sample. Results for Woman_T1 sample listed in Table 3, shows great color difference post aging for the red color as indicated by the high ΔE^* value. On the other hand, there is a slight color change for the following colors: yellow, purple, brown, and green.

Results for sample Woman_T1 reveal that the red color is the most affected by light aging and accordingly it appears lighter ($\Delta L^* = 6.57$) and more red ($\Delta a^* = 4.52$). On the other

hand, the purple color is the least affected by light aging as shown in Table 3. The quantities, with a uncertainty of 4%, shown in Table 3, were calculated at the end of the test aging (26 hours). The measurement points, listed in Table 3 and then in Table 4, were named with the code laboratory and Y, Bw, R, G, P mean respectively yellow, brown, red, green and purple.

Table 3. Average change in the three color coordinates and overall
color change for sample Woman_T1 after light aging

Measurement points	ΔL^*	Δa^*	Δb^*	ΔE^*
Y	2.92	-0.18	1.90	3.48
Bw	-0.39	2.75	2.18	3.52
R	6.57	4.52	1.25	8.07
G	0.88	0.41	-2.07	2.28
P	0.04	1.38	1.08	1.75

With regard to humid heat aging for sample Woman_T1, high ΔE^* values for all five selected colors purple, yellow, red and green are recorded. All values, shown in Table 4, were calculated at the end of aging (72 hours). In particular, results revealed that the yellow color is the most affected by humid heat aging and accordingly it appears lighter ($\Delta L^* = 5.49$), more yellow ($\Delta b^* = -1.72$), and more green ($\Delta a^* = -2.02$). Red is the second most affected color by humid heat aging and accordingly it appears less light ($\Delta L^* = -8.14$), and more yellow ($\Delta b^* = 15.56$). On the other hand, the green color is the least affected by humid heat aging.

Table 4. Average change in the three color coordinates and overall color
change for sample Woman_T1 post humid heat aging

	ΔL^*	Δa^*	Δb^*	ΔE^*
Y	5.49	-2.02	41.72	42.42
Bw	4.86	-9.94	27.3	29.47
R	-8.14	-32.47	15.56	37.36
G	6.51	-5.48	0.05	8.51
P	-15.70	- 20.01	16.48	30.31

Conclusion

The main aim of the research is to identify pigments used in hand-colored photographs, but also to study color change as a form of damage and define what causes it using non-destructive analysis; in order to select the most appropriate conditions to preserve such photographic materials and hopefully retard future damage. To achieve this aim, several analysis techniques were used including multispectral imaging, X-ray fluorescence spectroscopy and colorimetric analysis.

Multispectral imaging, in particular RGB and False Color IR, analyzes the photographs in RGB (red, green, blue) acquisitions for the high resolution documentation and false color Infrared. This technique gives the possibility to identify the components of pigments which are similar in color but different in composition.

Colorimetric analysis allows the characterization of all samples colors. The identification of the pigments was achieved using this technique through the comparison among the experimental and database SRF% trends.

Colorimetric analysis was necessary for quantifying the color changes before and after artificial aging, in particular, to UV and humid heat aging.

XRF analysis was used primarily to identify the photographic processing chemistry and the nuances associated with chemicals. It was also possible to identify others elements useful for the identification of the pigments (i.e. cinnabar, realgar, cobalt, and yellow ochre).

Artificial aging has been conducted on some samples to study the effect of light, and humid heat on hand-colored photographic prints. Results show that the ratio of color change in humid heat aging is higher than that of the light one. Some colors have been affected more than others when exposed to humid heat aging. This probably depends on the chemical composition of the pigments used.

Acknowledgments

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