

Evaluation of coated laser printed in fills for use in manual restoration of losses in silver gelatin prints

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Abstract

The significance of photographic collections to our shared cultural heritage has become apparent; and therefore, the focus on their preservation has intensified. Photographs made during the last one hundred years belong to a class known as silver gelatin prints. A silver gelatin print is mainly composed of the primary support (i.e. paper), the baryta layer (i.e. fine particles of barium sulfate in gelatin layer) and the gelatin binder layer carrying the final image material (i.e. metallic silver particles). One major issue in photograph conservation field repairing losses in silver gelatin prints since the reflective index and texture of silver gelatin prints are difficult to simulate. In 2017, a combined manual and digital restoration technique was proposed by Yosri for the restoration of losses in silver gelatin prints. Results proved the stability of long-term stability of laser printing compared to inkjet printing. However, analytical assessment techniques showed changes in the appearance of the samples post artificial ageing. The main aim of this paper is to assess the efficiency of selected coating materials (i.e. methyl cellulose, Klucel G, gelatin and Funori) in improving the stability of laser printed infills to be used in damaged silver gelatin prints. Long-term stability of the coated printed samples will be evaluated after exposure to humid heat artificial ageing at a temperature of 80°C and 65% RH for 5 days using several techniques including visual inspection, microscopic inspection, pH value measurements, colorimetric measurements, and Fourier transform infrared spectroscopy (FT-IR). The second part of the study involves the treatment of selected vintage silver gelatin prints suffering from various damage forms, mainly losses and surface dirt.

Keywords: Silver gelatin prints; losses; laser printing; digital restoration; manual restoration; accelerated ageing; consolidation; visual inspection; pH value measurement; colorimetric measurement; FTIR; conservation.

1. Introduction

In Egypt, libraries, archives, museums and research institutions house significant photographic collections. Photographic collections may be characterized as fine art or documentary [1]. Recently, the significance of photographic collections to our shared cultural heritage has become apparent; and therefore, the focus on their preservation has intensified [2]. Growing concern for the preservation of photograph collections has led to a greater interest in developing

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Appropriate conservation procedures to help prolong the lifespan of such valuable records. Photographs made during the last one hundred years belong to a class known as silver gelatin prints [3]. Several inventors, including: Peter Mawdsley, Josef Marieeder, Giusppe Pizzighell, and Sir William de Wiveleslie Abney, can be credited with the most important contributions to its development and research of several key types of silver halide gelatin emulsions [4], [5]. Silver gelatin prints were available both as POP (i.e. printed-out prints) and DOP (i.e. developed-out prints). The main difference between them lies in the way the silver-based image is formed [5].

A silver gelatin print is mainly composed of the primary support (i.e. paper), the baryta layer (i.e. fine particles of barium sulfate in gelatin layer) and the gelatin binder layer carrying the final image material

(i.e. metallic silver particles) [6], [7]. Due to the multiplicity and complicity of their structure, silver gelatin prints are extremely vulnerable towards their surrounding environment [8]. Factors that affect the permanence of photographic collections include inherent vice, improper temperature and relative humidity levels, improper light level, atmospheric pollutants, biological threats, poor display and storage conditions, inappropriate handling and disasters [9]. The resultant damage forms can be divided into the following categories according to their origin: physical, chemical, and biological damage as well as deposited matter [10], [11]. Deterioration may occur gradually or rapidly depending on the initial processing and subsequent keeping conditions and handling [12]. Physical forms of damage include all forms that originate by poor handling such as emulsion flaking, tears, scratches, losses, etc. On the other hand, chemical forms of damage originate from chemical reactions, and these include image fading, image discoloration, and binder and support discoloration and embrittlement, while biological damage includes all forms caused by living organisms [10], [13]. Each of the three layers that form a silver gelatin print is susceptible to specific types of damage [14]. The issue of deterioration of photographs presents special challenges to collectors, librarians, archivists and conservators. These outstanding visual records exist in relatively large numbers; and therefore, they are in urgent need of conservation treatment, rehousing, increased accessibility, and improved environmental conditions [15].

One major issue in photograph conservation field repairing losses in silver gelatin prints since the reflective index and texture of silver

gelatin prints are difficult to simulate [16]. The conventional method of restoration is to use a piece of paper that is similar to the original in weight, texture and color, on which the outline of the missing area is transferred using a pin tool. The edge is then chamfered using a scalpel and fixed in place using an appropriate adhesive such as carboxy methyl cellulose (i.e. CMC) or hydroxyl propyl cellulose (i.e. Klucel G) [16], [17]. Restoration of photographs has been going on since the birth of photography. Preservation of image information included practices such as retouching [18]. Lead pencils, carbon pencils and water colors are commonly used for photo retouching. Color is applied in the form of a series of minute dots, a technique known as stippling. Different binders have been added to retouching colors to obtain a gloss matching the original surface [16]. Large fills is traditionally carried out using an airbrush [16], [19], [20]. Nevertheless, there are some ethical issues involved in retouching photographs after in-filling treatments since all available retouching techniques modify the original surface characteristics in a way that its integrity could be compromised [16]. One of the common disadvantages of this practice is inhomogeneity that arises due to changes in the photographic image or the inpainting media or both. At times, skillfully applied original inpainting becomes unsightly when the surrounding image fades and discolors, leaving the retouched areas extremely obvious [21].

Digital restoration (i.e. digitization) is a method used for the virtual reconstruction of damaged and/or lost image information [18], [22]. Photograph conservators prefer this technique over manual restoration

since it does not involve any physical manipulation of the original photograph beyond making a high-quality scan. All restorative work is carried out on the computer which means there is much less risk of damage to the original than with conventional restoration processes. [23]. It is less time consuming compared to traditional restoration processes [18]. One important advantage of digital restoration is that the results are permanent unlike conventional restoration which is subject to damage [23]. For the previous reasons, the best option would be to digitally restore deteriorated photographs, but only when urgently needed, for display purpose where the flawless appearance of the displays is of great importance, this is not always the case, but conservators are faced with similar situations. The process of digital photo restoration begins with scanning the old photograph at a resolution that is high enough to capture all the image information [24]. Once the image is in a digital form, multiple image editing softwares can be employed to carry out the restoration work. Photoshop is the primary image editing software used since it contains powerful tools [10], [23], [24].

There are many printing technologies for output from computers; however, the most popular are inkjet and laser printing [25]. Inkjet printing is a material-conserving deposition technique used for inks. These inks consist of a solute dissolved or otherwise dispersed in a

solvent. The process essentially involves the ejection of a fixed quantity of ink in a chamber, from a nozzle through a sudden, quasi-adiabatic reduction of the chamber volume via piezoelectric action. A chamber filled with liquid is contracted in response to application of an external voltage. This sudden reduction sets up a shockwave in the liquid, which causes a liquid drop to eject from the nozzle [26]. On the other hand, Laser printing is based on the principle of electrophotography, where fine polymeric toner powder is picked up and precisely deposited on a substrate, usually paper, using electrostatic forces [27]. Digital prints are highly variable in their sensitivities to factors of deterioration [25].

In 2017, a combined manual and digital restoration technique was proposed by Yosri for the restoration of losses in silver gelatin prints. Results proved the stability of long-term stability of laser printing compared to inkjet printing. However, analytical assessment techniques showed changes in the appearance of the samples post artificial ageing [21].

The main aim of this paper is to assess the efficiency of selected coating materials (i.e. methyl cellulose, Klucel G, gelatin and Funori) in improving the stability of laser printed infills to be used in damaged silver gelatin prints. Long-term stability of the coated printed samples will be evaluated after exposure to humid heat artificial ageing at a

temperature of 80°C and 65% RH for 5 day using several techniques including visual inspection, microscopic inspection, pH value measurements, colorimetric measurements, and Fourier transform infrared spectroscopy (FT-IR). The second part of the study involves the treatment of selected vintage silver gelatin prints suffering from various damage forms, mainly losses and surface dirt.

2- Testing a dual manual and digital restoration technique

2.1 Materials and Methods

2.1.1 Materials

Three silver gelatin prints were selected for this study, three of which have losses among other damage forms. A 300 gm matt printing paper and HP Laser printer were used to prepare the infills. Four coating materials were prepared for this study: methylcellulose, Klucel G, gelatin and funori in three different concentrations (i.e. 1%, 2% and 2.5%) to assess their efficiency in stabilizing laser printed infills to be used in the manual restoration of deteriorated silver gelatin prints. Methylcellulose is the methyl ether of cellulose, prepared from wood pulp or cotton after treatment with alkali and methylating agent as methyl chloride or dimethyl sulfate. Methylcellulose is hygroscopic, odorless, fine granules, filaments or powder, swells in water, viscous, insoluble in ethanol, ether and chloroform [28]. Klucel G is grade of the cellulose ether, known as hydroxypropyl cellulose (HPC). Klucel G is soluble in water or polar solvents forming a gel or solution [29]. It is common in the conservation field as it is used for the consolidation of paper and leather, as well as a coating material [30],

[31], [32], [33]. Funori is a polysaccharide extracted from the red alga *Gloiopeltis furcata*, the seaweed is cleaned, soaked, pressed and dried to give thin sheets. The mucilage is extracted with hot water and the residual material filtered off. Funori is applied as a warm solution in water for consolidation purposes [34] Gelatin is produced in many different grades. Those recommended for use in conservation are "photographic quality" gelatins which are highly purified, and available from Eastman Kodak and Fisher Chemicals. Low grades of gelatin are not suitable for use in conservation as consolidants as they contain metallic salts and other impurities should be tested with mock-ups and locally on an object before using overall. It may be desirable to use repeated layers of thinner solutions of gelatin to avoid gloss problems [35]. Table 1 shows the samples, the concentrations used as well as the sample numbers.

Table 1: Samples prepared for this study

Materials	Materials Key	Sample Numbers		
Funori	F	F1%	F2%	F2.5%
Methylcellulose	MC	MC1%	MC2%	MC2.5%
Klucel G	K.G	K 1%	K2%	K2.5%
Gelatin	G	G1%	G2%	G2.5%

2.1.2 Methods

1. 2. 1. 1. Application Methods

1. 2. 1. 1. 1. Digital Restoration

The digital restoration was performed using Adobe Photoshop version cc 2017. The tools used to digitally restore the missing parts in the silver gelatin prints include: Spot Healing Brush tool (J) and Healing Brush tool (J) which replaces a selected area using a pattern or pixels from another part of the image; Clone stamp tool (S) which paints with pixels from another part of the image; Eyedropper tool (I) samples colors from an image; Brush tool (B) which paints custom brush strokes; and the Pen tool (P) which makes and changes paths or shapes with anchor points and handles

1.2. 1. 1. 2. Laser printing

After digital restoration, 300 gm matt printing paper was employed for printing the digitally restored areas using an HP Laser printer. The best results for the printed samples were selected [**Fig. 1**].



Fig. 1. The printed laser infills prepared for this study.

1. 2. 1. 1. 3. Coating Technique

The prepared coating material solutions were applied by brushing the solution on the digitally restored samples in one direction.

1. 2. 1. 2. Artificial Ageing

Treated samples were artificially aged at a temperature of 80°C and 65% RH for 5 days, which is equivalent to ageing of paper under normal conditions for 25 years. The ageing process was in conformance with the ISO 5630-3:1996 standard [36] [37]. This procedure was performed in a Binder dry oven with digital indicator, model no. 92403000002000 at the National Institute of Standards (NIS) in Cairo, Egypt.

1. 2. 1. 3. Investigation and Analysis Techniques

1. 2. 1. 3.1. Visual Inspection

Visual inspection was used to initially evaluate the tested coating materials based on the apparent changes that have occurred as a result of artificial ageing.

1. 2. 1. 3.2. Microscopic Inspection

This investigation tool was used to monitor the visual changes which have occurred due to artificial ageing. A ROHS Digital USB microscope 1000X was used.

1. 2. 1. 3. 3. Colorimetric Measurement

The change in color due to treatments was measured using MiniScan Model No. EZ MSEZ0693. All samples were measured in a visible region, with an interval of 10 nm using D65 light source and an observed angle of 10 degrees. The CIELAB color parameters (L^* , a^* , b^*) were used, where L^* defines lightness and varies from 0 (black) to 100 (white); a^* represents the red/green axis, where $+a$ means red and $-a$ means green; b^* represents the yellow/blue axis, where $+b$ means

yellow and -a means blue. All values of L*, a*, and b* were obtained before treatment and after treatment and artificial ageing. Each reading was the average of three measurements. The total color difference ΔE^* was also calculated from the following formula: $\Delta E^* = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2}$ [38] [39]. The analysis was carried out at the National Institute of Standards (NIS) in Cairo, Egypt.

1. 2. 1. 3. 4. Fourier Transform Infrared Spectroscopy (FT-IR)

FT-IR spectroscopy was used to study the chemical changes which may have occurred after treatment and artificial ageing. The FTIR instrument used is Nicolet 380 FT-IR Spectrometer under transmission mode. The analysis was carried out at the National Institute of Standards (NIS) in Cairo, Egypt.

2.2 Results and Discussion

2.2.1. Visual inspection

Post artificial ageing, no visible sign of color change in samples [G, F and MC] in all three concentrations was observed; however, samples treated with Klucel G in all three concentrations [1%, 2% and 2.5%] showed visible color change. Accordingly, the Klucel G samples were excluded from this study [Fig. 2]. Initial results of the accelerated ageing test revealed the stability of samples [F, G and MC].

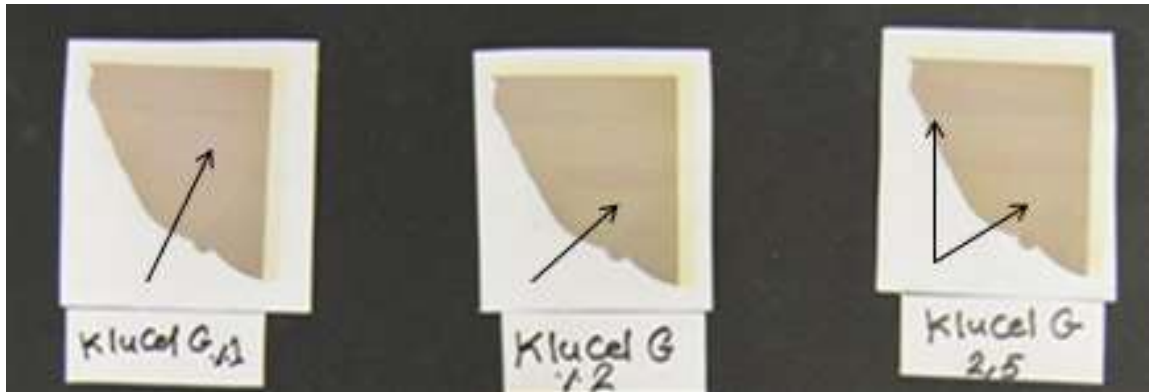


Fig. 2. Klucel G treated samples post artificial ageing showing visible color changes.

2.2.2. Microscopic inspection

Investigation by digital microscope showed no noticeable changes in all coated laser printed samples after accelerated ageing test [Fig. 3] [Fig. 4].

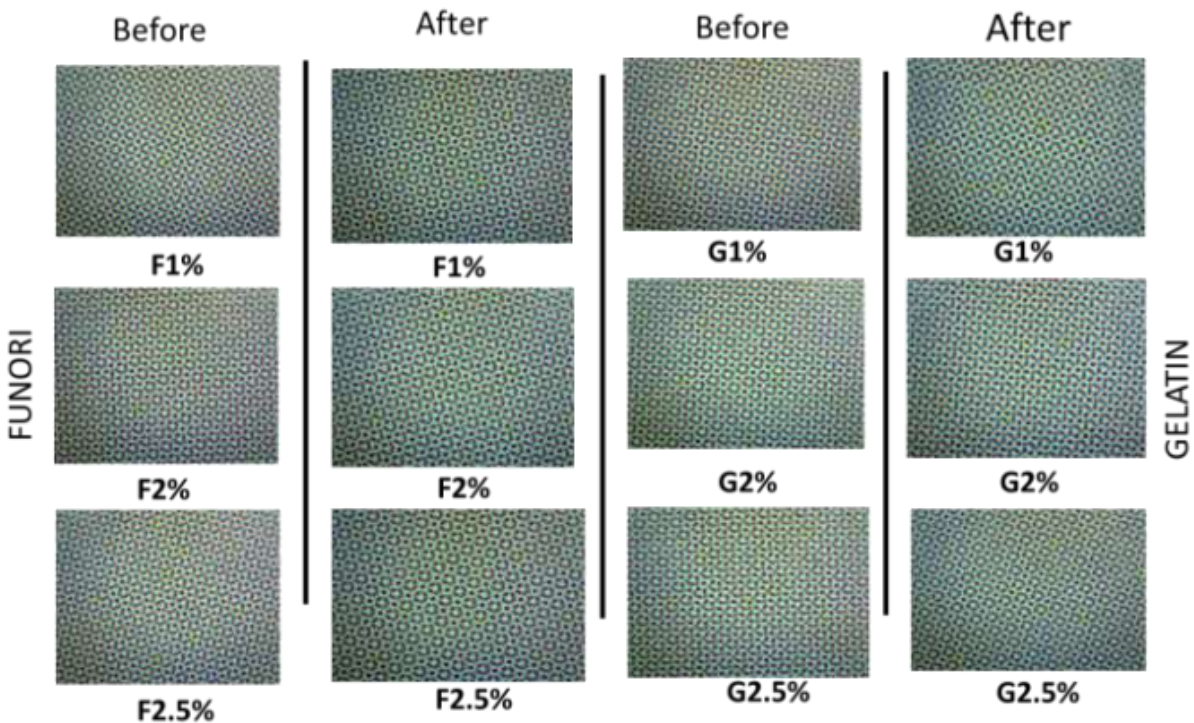


Fig. 3. Funori and gelatin treated samples before and post artificial ageing.

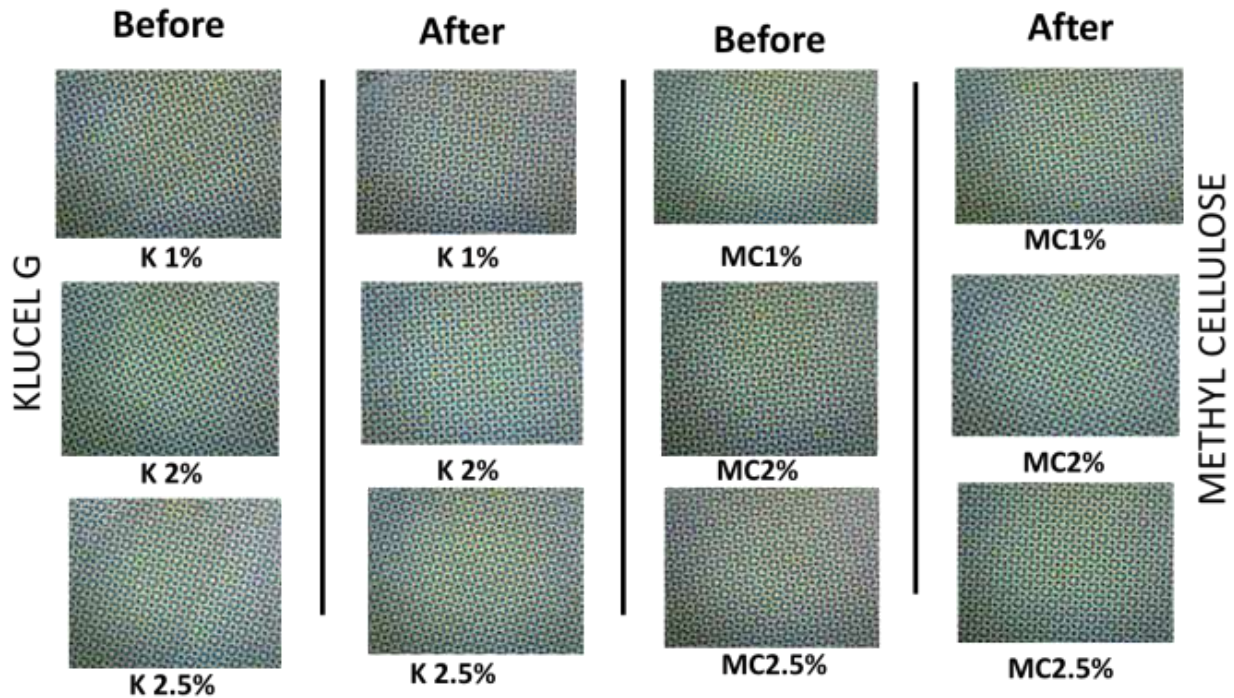


Fig. 4. Klucel G and methyl cellulose treated samples before and post artificial ageing.

2.2.3. Colorimetric Measurements

According to DIN EN ISO (super ceded by BS EN ISO 4628-1:2004), evaluation of ΔE^* is as follows:

- 0 – 1: color difference is not visible.
- 1-3: few people can recognize the difference.
- 3- 5: 66 % of people can recognize the difference.
- >5: everyone can recognize the difference [40], [41].

In literature, ΔE^* values of 2-3 are thought to be observable and unacceptable color difference [42]; however, it is clearly lower than the threshold limit ($\Delta E^* = 5$) required for the maintenance and restoration of historical surfaces [43].

Another study also mentions that a $\Delta E^* \ll 4$ is a value normally accepted as limit for the visual impact of surface treatments [44].

Results show that Klucel E has caused a visible change in the measured sample in some cases reaching a ΔE^* value above 5 [Fig. 5].

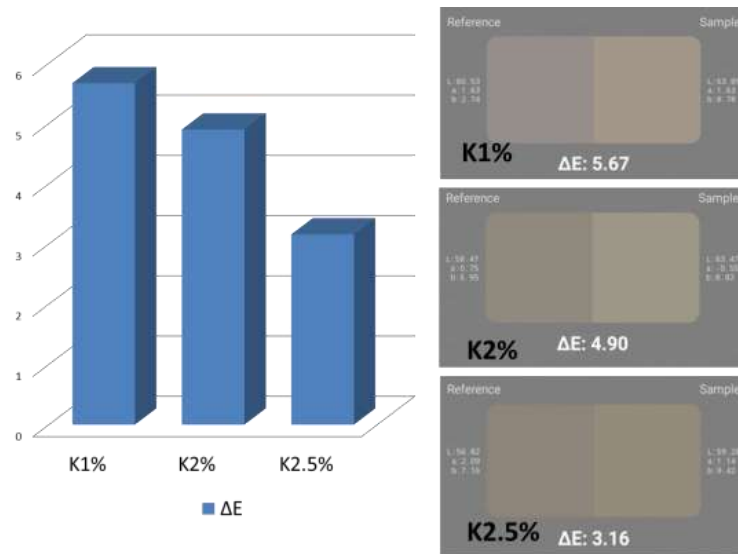


Fig. 5. ΔE^* values for Klucel G treated samples as a result of artificial ageing.

Funori showed ΔE^* values below 5 in all three concentrations [Fig. 6], which according to the international standards is considered acceptable as a treatment. On the other hand, gelatin gave good results excluding concentration 2% which gave a ΔE^* value above 6 [Fig. 7]. Methyl cellulose gave ΔE^* values below 5 for all tested concentrations [Fig. 8].

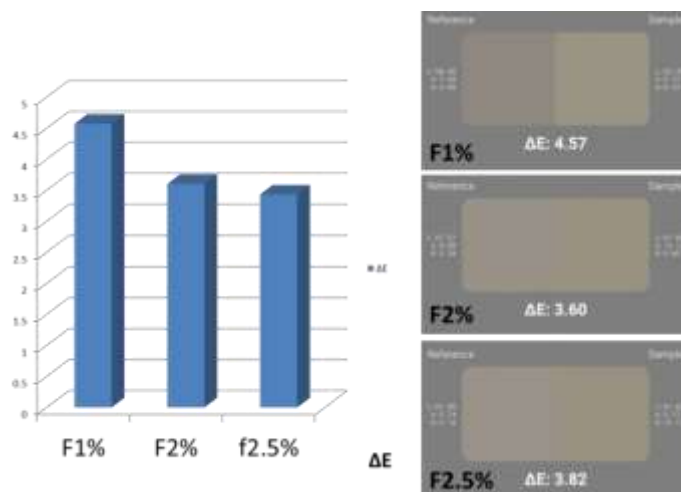


Fig. 6. ΔE^* values for Funori treated samples as a result of artificial ageing.

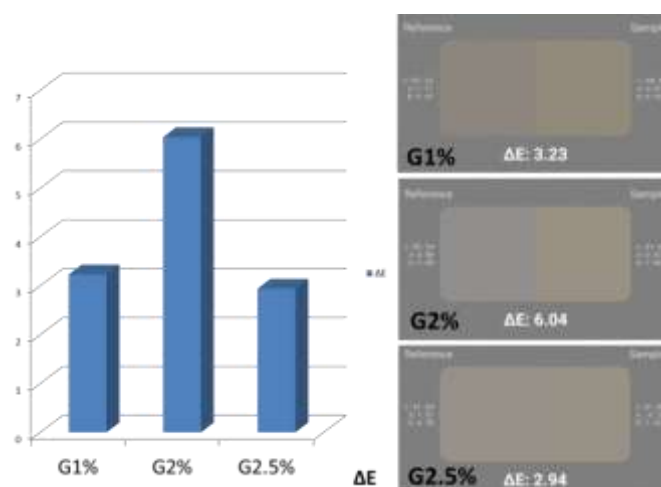


Fig. 7. ΔE^* values for gelatin treated samples as a result of artificial ageing.

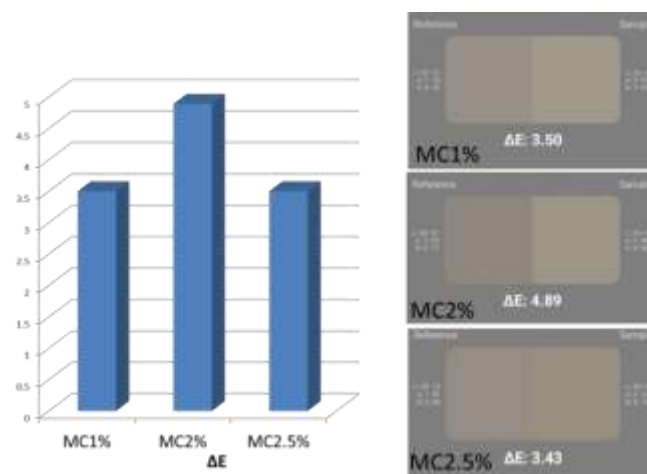


Fig. 8. ΔE^* values for methyl cellulose treated samples as a result of artificial ageing.

In breaking down the data L^* , a^* , b^* values, minor changes are observed in all three parameters when compared to the before aging samples. However, there is an increase in the b^* value after aging for the sample treated with Funori (2.5%), which indicated the occurrence of yellowing. Similarly, in all other treated samples, there is an increase in the b^* value in higher concentrations [Fig. 9].

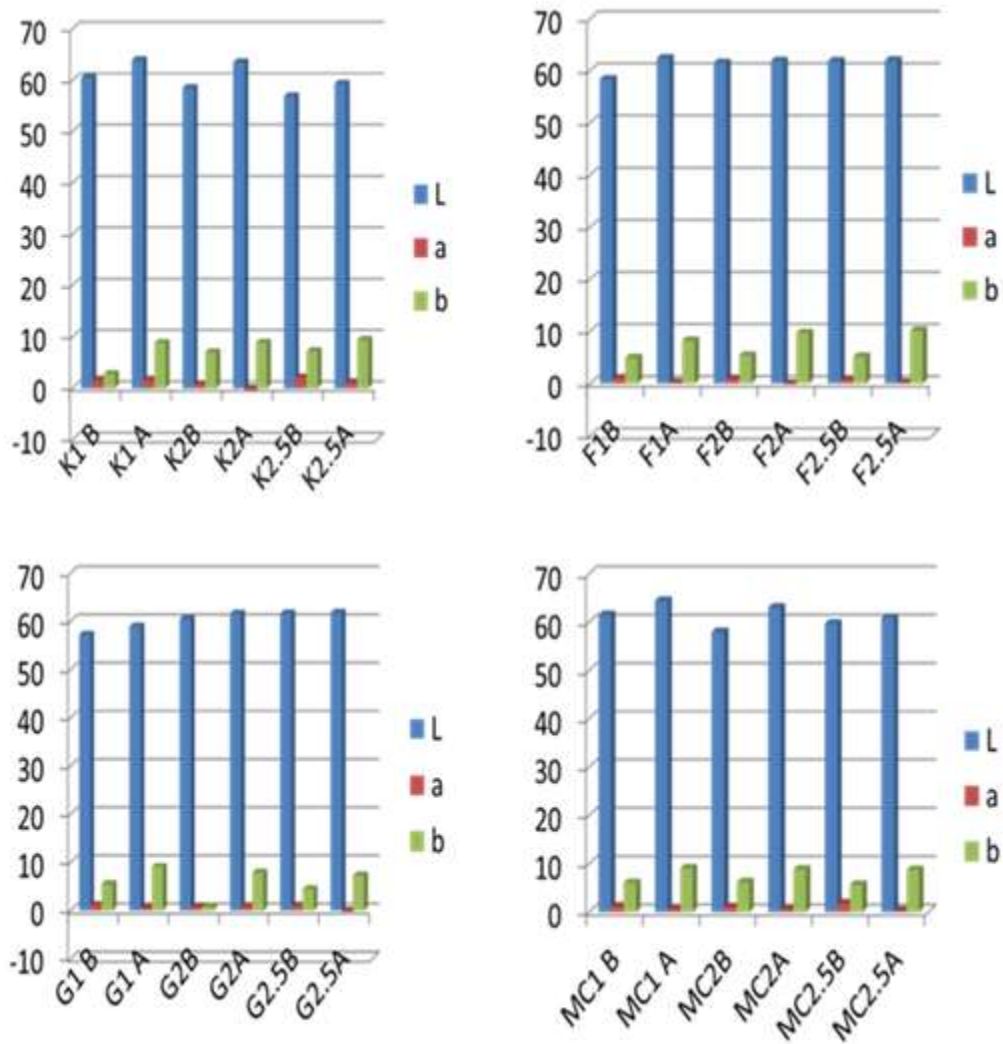


Fig. 9. L^* , a^* and b^* values for all treated samples before and artificial ageing

2.2.3. Fourier Transform Infrared Spectroscopy (FT-IR)

FT-IR results show no chemical changes of cellulose in all three samples. The carbonyl group at $1650-1698\text{ cm}^{-1}$, which is used as an indicator for oxidation has not been detected post ageing [**Fig.10**] [**Fig.11**] [**Fig.12**].

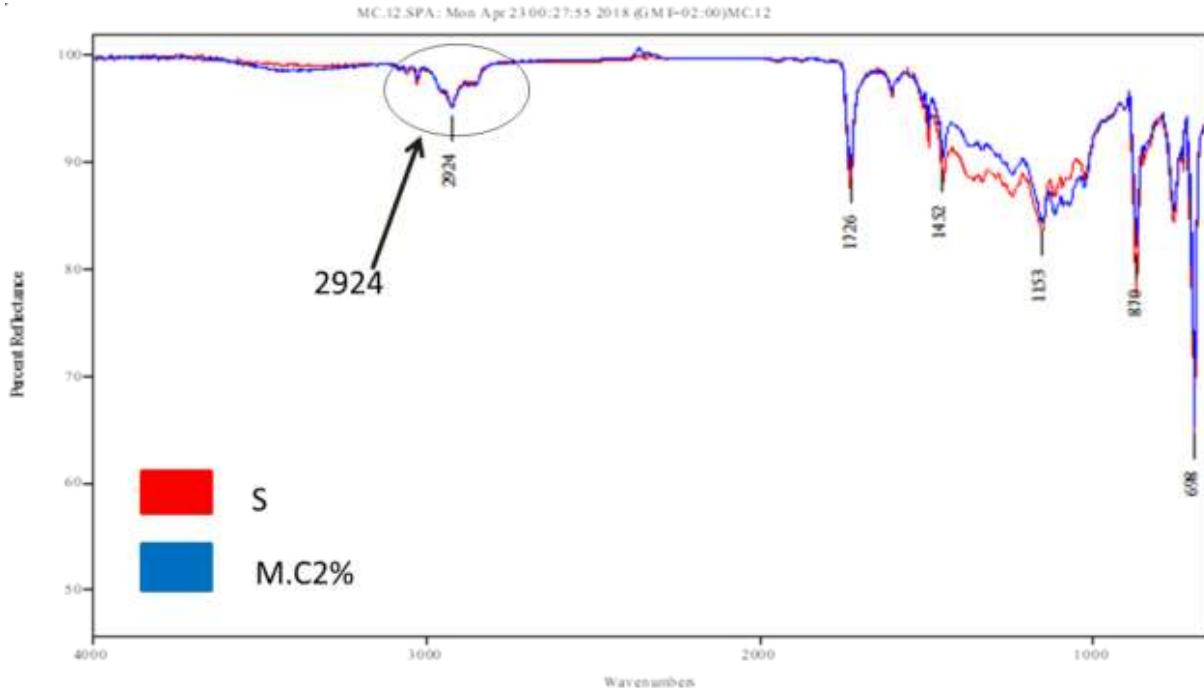


Fig. 10. FT-IR spectra for sample treated with 2% solution of methyl cellulose in distilled water before and after artificial ageing.

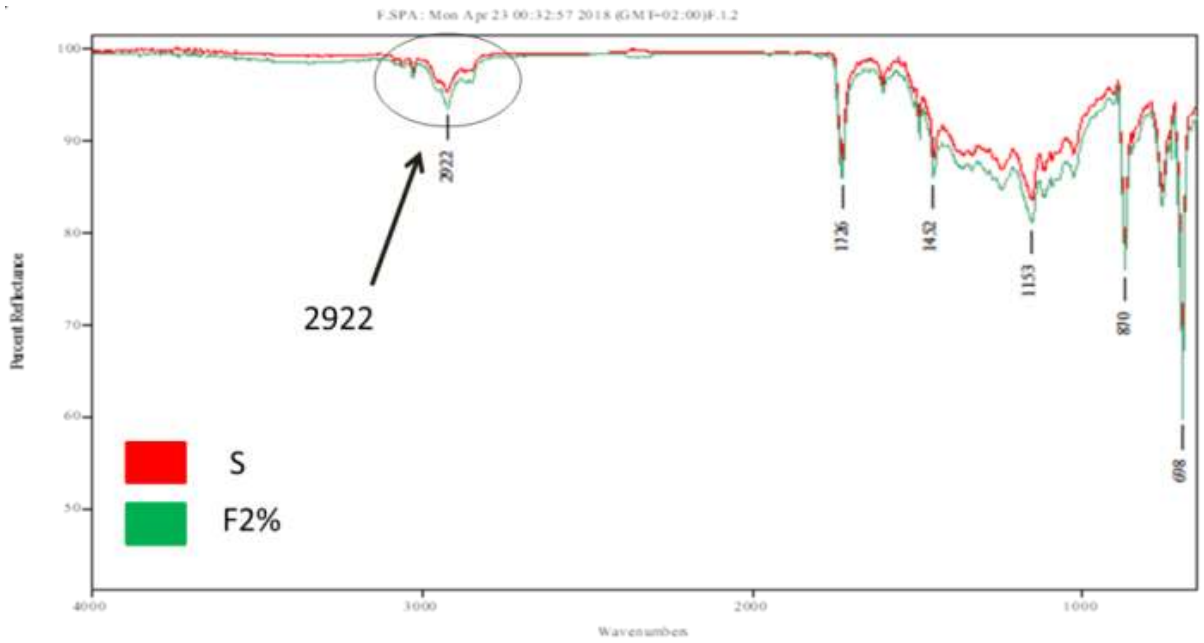


Fig. 11. FT-IR spectra for sample treated with 2% solution of Funori in distilled water before and after artificial ageing.

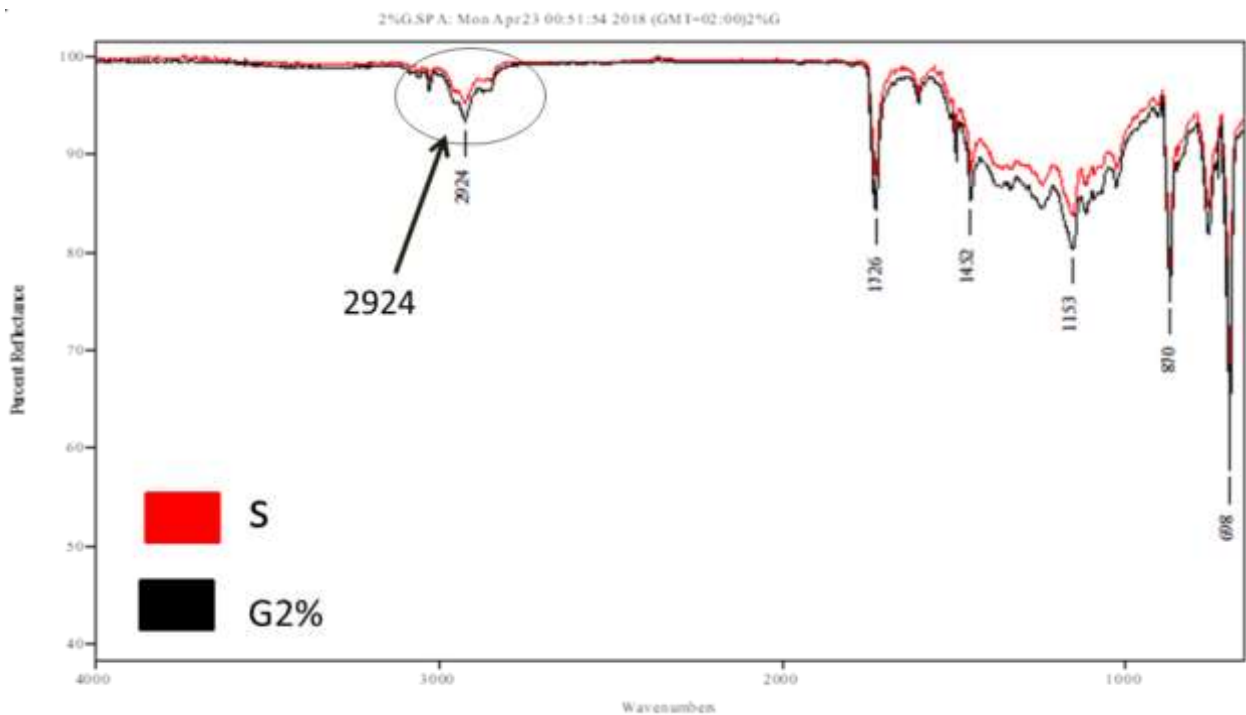


Fig. 12. FT-IR spectra for sample treated with 2% solution of gelatin in distilled water before and after artificial ageing.

3. Conservation of Three 20th Century Silver Gelatin Prints

3.1 Collection Description

The collection consists of three black and white photographic prints. One of the photographs is of an old lady sitting on a chair. The other two are of policemen; apparently both were taken during a ceremony. The lady portrait is framed in a black wood frame. The photos are of varying sizes. The photographic process has been identified as silver gelatin process due to the black and white image tones, the absence of paper fibers and the presence of silver mirroring as a common image silver decay form in silver gelatin prints collections [45], [46].

3.2 Condition Assessment

The general state of preservation for all three prints is good in term of its physical condition; however several forms have been recognized.

3.2.1. Visual Inspection

The two policemen photograph suffered from fingerprint stains, curling, stains, dust, silver mirroring and loss of corner, in addition to the presence of cracks which revealed the presence of the baryta layer distinguishing silver gelatin prints [Fig 13]. Second policemen group photograph suffered from the same damage forms as the first one (i.e. finger print stains, curling, stains, dust and silver mirroring and two losses in the edges [Fig. 14]. The lady portrait suffered from flaking, dust, stains and scratches [Fig 15]. Its wood frame of photo suffered from flaking, presence of microorganisms, remains of rust stains resulting from the metal nails. The cardboard paper board suffered from severe yellow, an indication of high acidity [Fig. 16].



Fig. 13. Damage forms found in photograph no. 1.

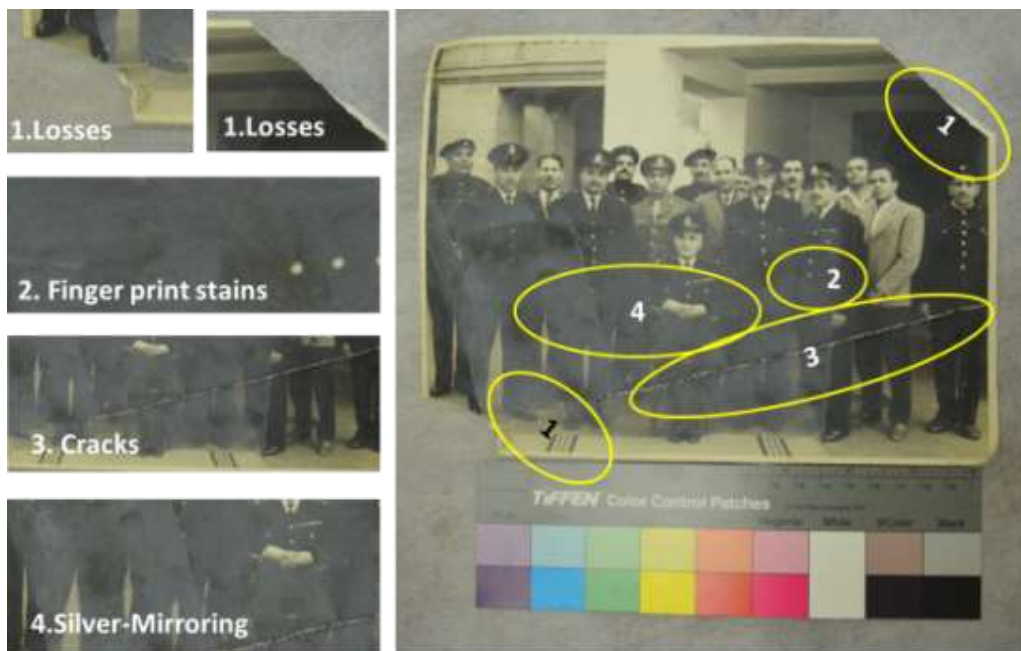


Fig. 14. Damage forms found in photograph no. 2.

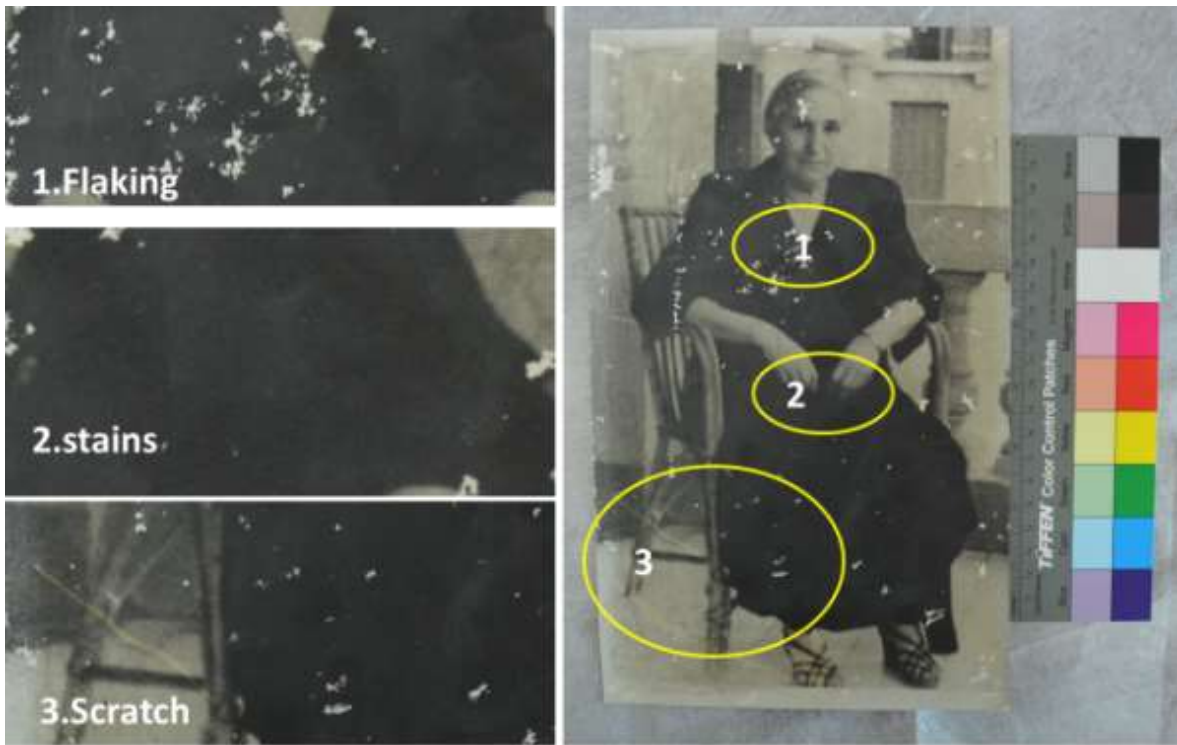


Fig. 15. Damage forms found in photograph no. 3.

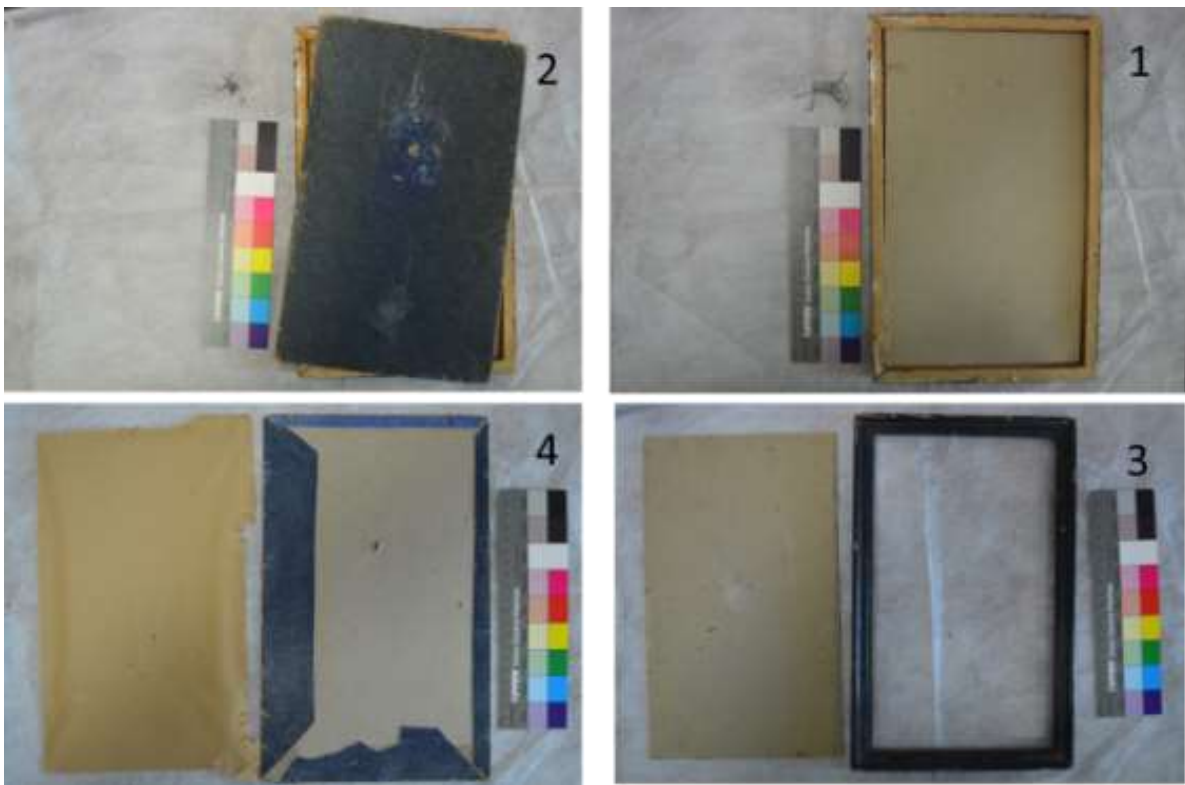


Fig. 16. Damage forms found in the components of the wood framing system used to house photograph no. 3.

3.2.1. Microscopic Inspection

ROHS Digital USB microscope 1000X was used to record deterioration forms found in this study [Fig. 17]. It was further used to document the efficiency of the used cleaning treatment, particularly the removal of silver mirroring.

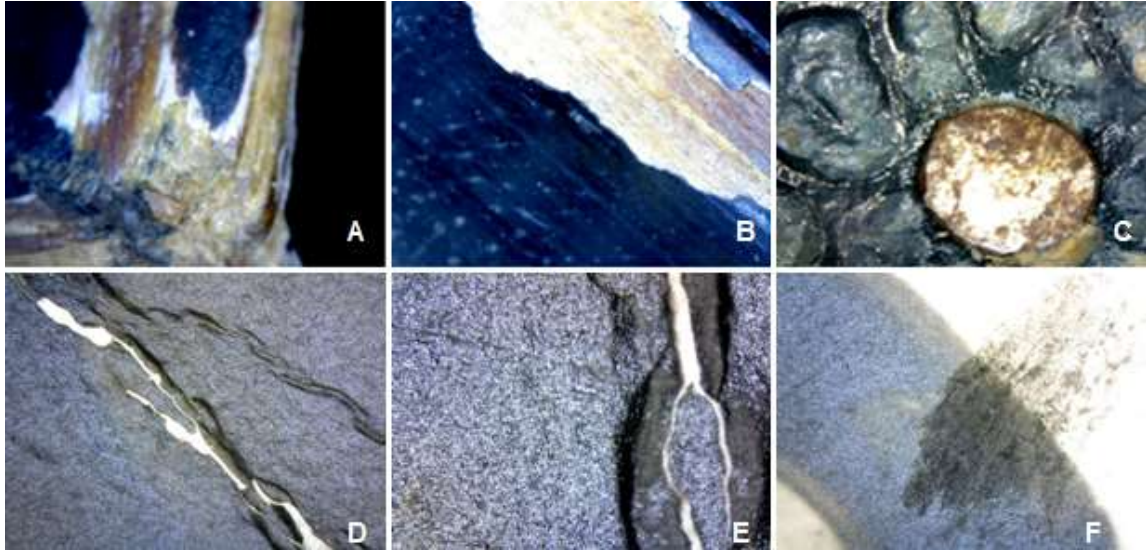


Fig. 17. Microscopic inspection images of the damage forms found in the photographic collection under study. (A) and (B) separation of the plaster , (C) rusted pin, (D) cracks, (E) silver mirroring, and (F) stains.

3.3. Conservation Interventions

3.3.1. Cleaning Process



Fig. 18. Mechanical cleaning of the photographic prints using vinyl erasers and fine brushes.

Mechanical cleaning was performed using several tools including scalpel, soft hair brushes, vinyl eraser and cotton to remove superficial dirt [Fig. 18]. Silver mirroring, fingerprints and some stains were removed using vinyl eraser and soft brushes to dispose harmful eraser residues [Fig. 19]. Chemical cleaning, organic solvent cleaning, was carried out using a mixture of 80% of ethyl alcohol in 20% distilled water to remove the remaining dirt and stains. A mixture of 50% of ethyl alcohol in 50% distilled water was used for the third photograph. The cleaning procedure also included chemical cleaning of the wood frame [Fig. 20].



Fig. 19. Microscopic inspection of silver mirroring removal using vinyl eraser.



Fig. 20. Solvent cleaning of photograph no. 3 (left) and mechanical and solvent cleaning of the wood frame using a scalpel, cotton swap and poultice technique.

3.3.2. Humidification

The photographic prints suffered from embrittlement, which caused the formation of cracks in two photographs. Accordingly, a humidification process was performed to restore their moisture contents. A sealed box was used for this treatment. Water at a temperature of 60°C was poured into two beakers, which were then placed inside the sealed box. Photographs were exposed to the water vapors for 10 minutes [Fig. 21].



Fig. 21. Humidification process performed to restore the moisture content of the brittle photographic prints.

3.3.3. Retouching

The frame of third photo suffered from flaking causing loss of frame color. Therefore, minor retouching was carried out using acrylic color which was applied by a fine brush [Fig. 22].



Fig. 22. Frame retouching with acrylic colors.

3.3.4. Compensating for losses

Laser printed infills coated with a 2% solution of Funori in distilled water was used to restore losses in the policemen photographs. The infills were secured in place using a Klucel G (i.e. 5% in distilled water) [Fig. 23] [Fig. 24].



Fig. 23. Compensation for losses using the proposed dual digital/manual restoration technique.



Fig. 24. Compensation for losses using the proposed dual digital/manual restoration technique

3.3.5. Display and Storage

The policemen photographs were preserved in suitable custom-made enclosure for future protection from sudden changes in temperature and humidity. Enclosures were made from acid-free paper made from pure cotton cellulose and polyester to minimize handling and to protect the photographs from light damage, dust, etc... Paraloid B-72 isolated nails were used to fix the backing cardboard to the wood frame [**Fig. 25**]. The collection should be ideally stored in cool, dry environment with good air and avoiding long term display.



Fig. 25. Enclosures for housing the collection.

4 . Conclusions

The investigation and analysis methods used in the paper proved the efficiency of digital restoration as a method for losses compensation. It further showed that the proposed digital/manual restoration technique is a very promising approach in the photograph conservation field.

Obtained results for the tested coating materials showed that Funori is suitable for use as a coating material with laser-printed infills in terms of preserving the optical and chemical properties of the infills as it showed no to insignificant change in color and no chemical change was detected post artificial aging.

Generally, the care of photographic collection depends on various factors including; creation of a controlled surrounding environment, the use of proper storage or display methods and materials.

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