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A Methodological Approach for Multi-Temporal Tracking of Silver Tarnishing

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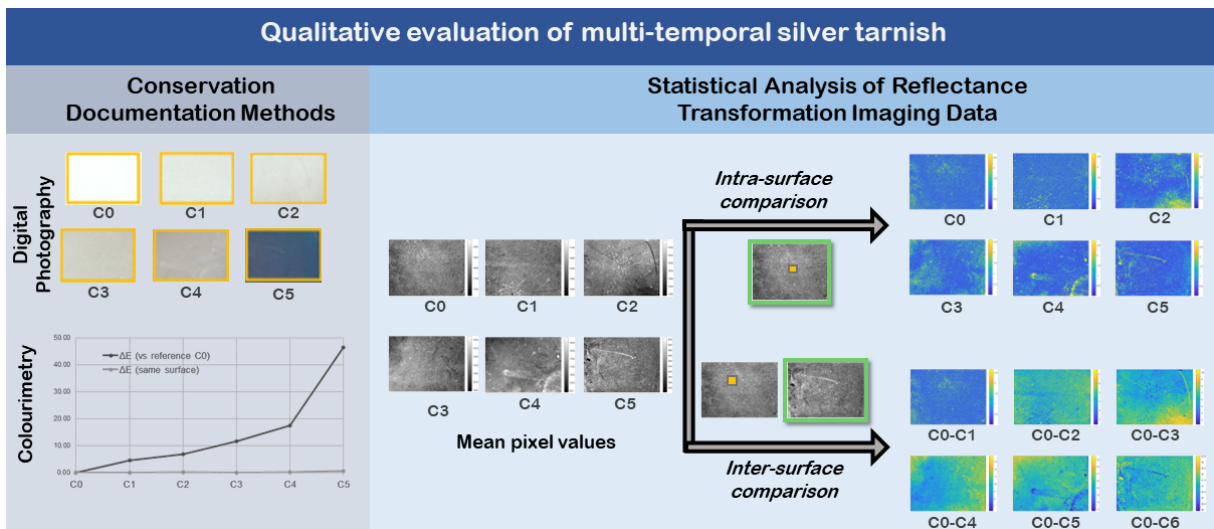
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ABSTRACT

Silver tarnish manifests by changes in the optical properties of the material. Documenting these changes creates many challenges for imaging techniques. This paper proposes a methodological approach based on processing Reflectance Transformation Imaging (RTI) data for tracking multi-temporal changes on such surfaces. Through the statistical analysis of the surface's angular reflectance, information related to the appearance attributes can be evaluated and visualized by maps. Thus, this paper explores the global surface change of the reflectance response of silver tarnishing as a function of time. A qualitative and semi-quantitative evaluation is based on multivariate distance measurements at different time intervals. The results are compared to surface change evaluation by visual inspection, photographic documentation, and colourimetry, practices traditionally used in conservation documentation to monitor surface changes over time. The outcome of this research illustrates the possibilities of RTI data analysis as a tool for accurate multi-temporal documentation of the optical properties changes on specular surfaces.

CCS CONCEPTS

- **General and reference** → **General conference proceedings**;
- **Applied computing**;

KEYWORDS

multi-temporal data, colourimetry, reflectance transformation imaging, silver tarnishing

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1 INTRODUCTION

Silver is considered a noble metal; nevertheless, it is susceptible to corrosion by atmospheric sulphur compounds creating a corrosion layer called tarnish [1–3]. Tarnishing is a slow process that alters the chemical composition of the surface due to the development of silver sulfide (Ag_2S) crystals [3–6]. These crystals form a passive film known for altering the surface's optical properties due to thin-film interference, which manifests as colour change ranging from a yellowish layer and extending to dark colours [1, 7].

Monitoring this gradual process is usually performed directly on objects by visual observation, photographic documentation, chemical or electrochemical analysis coupled with chemical imaging, or indirectly by evaluating the environmental conditions using silver sensors. Nonetheless, each method has advantages and disadvantages.

Environmental measurements can analyse the phenomenon that will cause the corrosion, but even when using silver as a sensor [8–10], it is an indirect measurement that cannot provide information about the changes in the object's surface. On the other hand,

the appropriate chemical/electrochemical analysis can give accurate information about the chemical composition of the corrosion products, measure the thickness of their layers and visualise the surface's chemical alterations at different magnifications [2, 3, 6, 11–13]. Nevertheless, many of these analyses can be destructive or alter the appearance of the metal surface and require specialised equipment. Finally, visual observation by experts (curators, conservator-restorers) is an invaluable tool for evaluating the surfaces based on perception and understanding of signs of corrosion development; yet, detecting minor changes at different time intervals is challenging. Similarly, imaging techniques can provide visualisations under different conditions, magnifications, and for different time intervals, but, to the authors' knowledge, there have been no reports on their systematic use of following changes related to silver tarnishing.

This paper aims to explore statistical data processing of image stacks acquired with Reflectance Transformation Imaging (RTI) as means of documenting the surface's optical properties in a non-invasive and accurate way. Test samples (coupons) were aged at different time intervals (Section 2). The resulting coupons were then evaluated using standard conservation-restoration monitoring practices through visual observation, photographic documentation, and colourimetry (Section 3). The proposed methodology for statistical processing of RTI data was then examined to provide an accurate qualitative analysis of surface alteration (Section 4). The advantages and disadvantages compared to traditional techniques are then discussed (Section 5). Preliminary results demonstrate the potentiality of using RTI statistical data processing to accurately evaluate surface change over time (Section 6).

2 COUPONS PREPARATION

A set of test samples (coupons) were artificially corroded and examined in terms of multi-temporal change detection of the surface optical properties. Silver sheets of size $4 \times 6 \times 0.2\text{cm}$, made of pure silver (Ag 99,9%) were selected to evaluate the effect of tarnish without the influence of alloying elements. Flat surfaces without decorations were selected to avoid interference with the results due to local textural changes or shape shading. First, the surfaces were sand-blasted to create an isotropic surface that is more adaptive to studying the effect of light reflectance with RTI. Next, they were cleaned electrochemically and degreased in an ultrasonic acetone bath before being artificially tarnished.

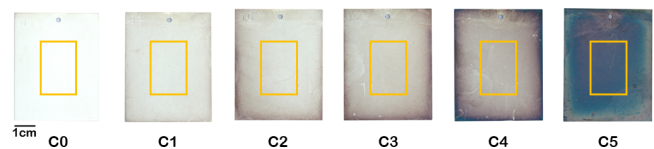


Figure 1: The different degrees of tarnish achieved on the silver coupons for the different time intervals. The square in the middle indicates the area of interest used for the RTI data processing.

Tarnishing was achieved by a protocol based on hydrogen sulfide (H_2S) vapours deriving from egg white powder (albumin) that enables the gradual creation of silver sulfide (Ag_2S) on the metal

Table 1: Silver test samples (Coupons)

Coupon Number	Hours of tarnish	Description
C0	00h	Reference
C1	02h	Tarnish level 1
C2	03.5h	Tarnish level 2
C3	05h	Tarnish level 3
C4	08h	Tarnish level 4
C5	12h	Tarnish level 5

surface. The protocol consists of creating an environment of hydrogen sulfide (H_2S) gas that is homogeneously distributed to the metal surface. For this purpose, 20%w/v of commercially available albumin in deionised water was used in a closed container (2.6L) with the coupons suspended over the surface while applying heat (80°C). The coupons were suspended in the middle of the container keeping a distance of more than 8cm from the albumin solution to ensure a homogeneous airflow. The process was repeated for the different time intervals per coupon resulting in a constant ratio of metal surface versus air for all degrees of tarnishing. This has created different tarnish levels that exhibit a gradual change in the optical properties of the surface. For each test sample, a photo, colour measurements and an RTI acquisition were acquired per time interval. The resulting coupons are presented in figure 1) and table1.

The tarnish layer develops as a chemical degradation that affects the entire surface; therefore, the experimental methodology aimed to evaluate the surface's average change for proposing a protocol for assessing the global surface change. Nonetheless, it must be noted that due to the edge effect (accelerated corrosion at the edges of the coupons due to differential aeration), an area of 1cm around the coupon was not considered in the surface evaluation.

3 APPLICATION OF CONSERVATION DOCUMENTATION METHODS

3.1 Visual inspection and photography

The first step of the evaluation was the visual inspection by a conservation-restoration expert. Next, technical photography of the coupons was performed in standardised conditions. First, photos were taken in RAW format with a DSLR camera in a lightbox with diffused light; then, the colours were corrected using a colour-checker (x-rite®). From the visual inspection, it is evident that the colour difference is well visible, especially when placing one coupon next to the other and comparing them with the reference C0. Nevertheless, it is difficult to ignore the edge effect that highly affects surface visualisation. There is a gradual colour change towards darker colours between C0 and C4 and a rapid shift between C4 and C5, where the colour shifts to dark blue from a brownish layer. Nonetheless, the minor difference between short consecutive time intervals, i.e. between C1 and C2, is not well distinguishable by the human eye (fig. 1). The RGB images do not provide further information from the visual inspection but facilitate surface documentation.

3.2 Colourimetry

Colourimetry has been developed as a technique that helps specify colours by assigning a numerical description for each colour, even viewed and illuminated under different conditions. It has since been used to specify and compare colour values through the CIELAB standard colourimetric observer. In cultural heritage, colourimetry has found application in different cases [14], whereas some limitations are discussed in [15]. In regards to evaluating objects made of silver, colourimetry has been used to evaluate cleaning treatments [6], the ageing of varnishes [16] and evaluating silver chloride films photodegradation [17]. Concerning evaluating silver tarnish over time, the potentiality of systematic monitoring of the surface change of the optical properties of silver by measuring the colour difference has been demonstrated in [2, 4].

Colourimetric measurements were performed with a spectrophotometer. These devices measure the reflectance from a surface as a function of wavelength. An X-rite, Pantone, CI62, sphere spectrophotometer with a D65/10 illuminant and a sampling size of 0.8mm was used. In addition, SPIN (Specular Included) measurements were selected to include any change in lightness due to specular reflection. The resulting values are the average value of 5 measurements on different areas of the same surface within the ROI. Data were collected in L^* , a^* , b^* values for each time interval and represented graphically to visualise the shift of the colour change over time. In addition, a distance measurement between the reference and different time intervals was calculated for the change in lightness (ΔL^*) and the overall colour difference (ΔE).

$$\Delta L^* = L_t^* - L_{ref}^*, \quad (1)$$

$$\Delta E = \sqrt{(L_t^* - L_{ref}^*)^2 + (b_t^* - b_{ref}^*)^2 + (a_t^* - a_{ref}^*)^2}, \quad (2)$$

where ref corresponds to time zero and $t = [1, 2, 3, \dots, n]$ to the different time intervals.

The results of the colour measurements are presented in figure 2. The coupons are represented by their actual colour based on [18], which provides 3D and 2D L^* , a^* , b^* plots. These graphs help visualise the surface's colour shift. The most intense change is observed in the axes L^* and b^* where, over time, there is a gradual decrease of the lightness (the surface becomes darker) and an initial positive shift in the b^* for coupons C0 to C4, towards yellow colours, with a sharp change to negative values, blueish hues, for C5.

Based on the above information, the overall colour change over time was measured with the ΔE and ΔL^* . A constant reference was used to compare results, which in this case was the reference coupon (C0). An initial colour distance measurement of the reference (C0-C0) was calculated for both ΔE and ΔL^* to evaluate the system error and the resulting value was zero (0).

Overall, the spectrophotometer can accurately measure the colour difference even at levels not perceived by visual observation (i.e. $\Delta E(C1, C2) = 2.3$), whereas the ΔE and ΔL^* measurements provide a semi-quantitative measurement of the resulting change. It becomes evident that the colour change is relevant to the tarnishing time. Nevertheless, colourimetry estimates only the colour change of the surface without providing information on the geometric

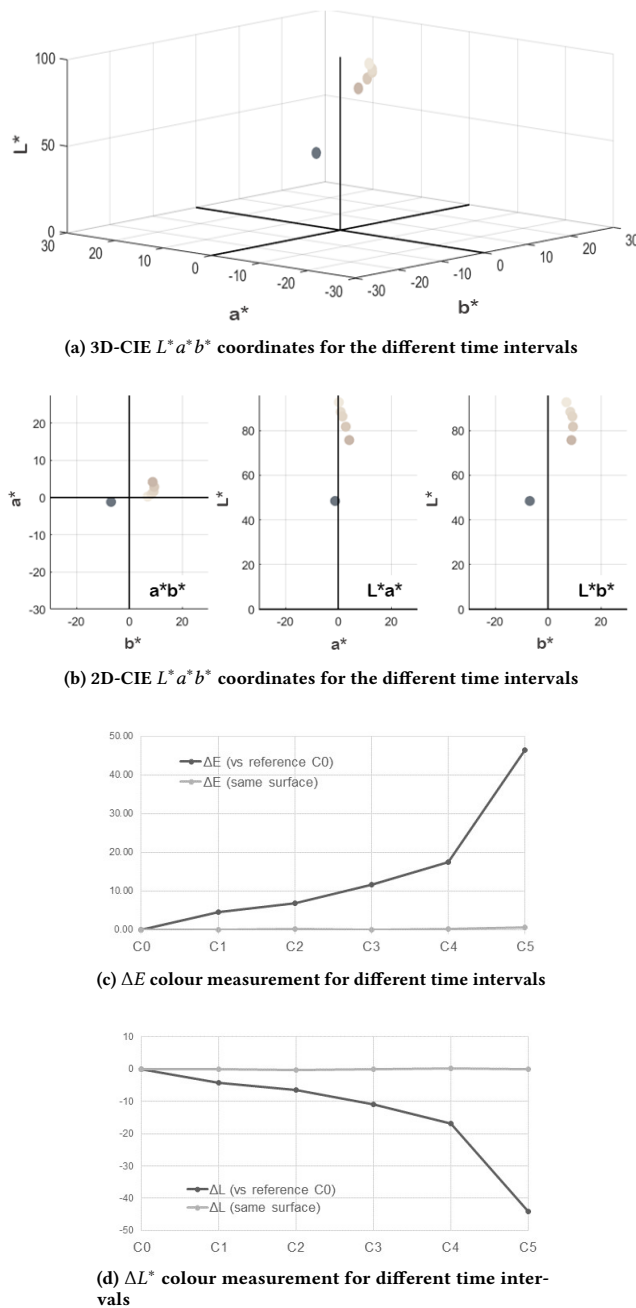


Figure 2: Multi-temporal colourimetry measurements demonstrate the color change of the metal coupons at the different time intervals.

attributes of the surface. Furthermore, the surface needs to have physical contact with the instrument, to ensure that no external light enters the illumination sphere during measurement. This can

alter soft metal surfaces like silver and make the method unreliable for curved objects. Finally, it lacks spatial localisation of the resulting change since no visualisation is possible.

4 PROPOSED METHODOLOGY

4.1 Reflectance Transformation Imaging

Reflectance Transformation Imaging (RTI) is a Multi-Light Image Collection (MLIC) technique that has found interest in cultural heritage applications since it can provide enhanced visualisation of the micro-geometric characteristics of a surface. These visual enhancements provide essential information on the surface's micro-geometry that helps better visualise inscriptions, surface anomalies, and decorative or technological characteristics of a surface with application to different materials [19–23]. RTI has also been exploited, by several research groups, as a tool for conservation documentation, mainly related to documenting physical damage, cleaning or other restoration treatments [24–28].

To the authors' knowledge, references to monitoring conservation treatments, or other types of multi-temporal data, focus on the geometric changes of the surface that are visible by inspection. However, in the case of silver tarnish, the development of corrosion crystals occurs at the nanometric level, meaning that the surface micro-geometry is not visible by the human eye. Therefore, this research aims to evaluate the RTI data processing by statistical analysis of the per-pixel angular light reflectance on surfaces without significant geometric alterations between different time intervals. The goal is to provide a systematic method for non-contact global change tracking surface alterations that do not present visible changes in their surface geometry. This methodology can have application as a preventive measure in the systematic monitoring of silver collections.

4.2 Methodological Approach

The general principle consists of creating an image stack from varying light positions keeping a constant distance between light source and surface. The method requires that both the surface and the camera are in fixed positions, and only the light changes its position [29, 30] (3). The resulting image stack can be exploited in different ways [26, 27, 31–35].

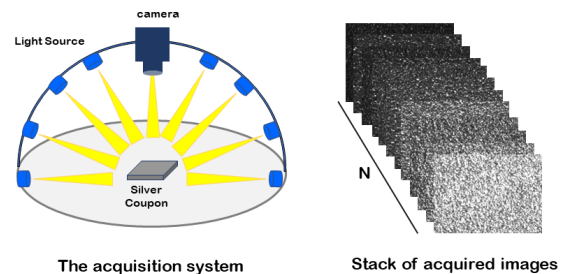


Figure 3: Simplified presentation of the acquisition system and the acquired image stack

In this paper, statistical analysis is based on calculating or comparing the mean per-pixel angular reflectance response deriving from the raw RTI data in the following ways: (fig. 4) [34]:

- **Feature Maps:** Correspond to the per-pixel mean of the reflectance response in the image stack for the different light positions (lp) (eq.4).
- **Intra-surface Distance Maps:** Compare each pixel's mean value with the mean of the entire surface of the feature map by applying the Mahalanobis Distance (eq.3,5).
- **Inter-surface Distance Maps:** Calculate the mean per-pixel distance of an image stack to the mean of a different mean feature map, again by applying the Mahalanobis Distance (eq.3,6). In this case, one mean feature map is considered the reference and the other mean feature map are the sample images.

Mahalanobis distance (MD) is a multivariate statistical measure of a point over a distribution, allowing for the evaluation of two variables from a centroid [36]. For RTI data, the point is a pixel, and the distribution is the entire set of pixels of the imaged surface. Furthermore, this distance is unitless and scale-invariant, aiding in a global surface evaluation of multiple variables.

$$MD_x = \sqrt{(x - \mu_y)\Sigma^{-1}(x - \mu_y)'}, \quad (3)$$

where x is the point to be compared and μ_y is the mean value of the entire data set (variables) and Σ is the inverse covariance matrix of the variables (y).

For statistical analysis of the raw RTI data, let's consider $I = [I_1, I_2, I_3, \dots, I_N]$ as a vector of the angular luminance for each pixel of an N set of images (e.g. red square denoted in the per-pixel raw statistical analysis 4).

The mean of each pixel is:

$$\mu(I) = \frac{1}{N} \sum_{k=1}^N I_k \quad (4)$$

This provides a new vector (M) of the mean per-pixel values that are visualized in a *mean feature map*, thus providing a general description of the surface appearance attributes (fig.4).

To calculate the MD for the intra-surface comparison eq.3 takes the following form:

$$MD_J = \sqrt{(J - \mu_M)\Sigma^{-1}(J - \mu_M)'}, \quad (5)$$

where J is the mean of the pixels ($\mu(I)$), μ_M is the mean of the mean feature of the other pixels, and Σ is the inverse covariance matrix of M . This creates a new vector M_1 representing the MD of each pixel in comparison to the mean feature, providing a visualization of the salient characteristics of the surface and allowing to set the lower threshold of detectable changes in the proposed configuration (fig.4).

Equivalently, to calculate the MD for the inter-surface comparison eq.3 takes the following form:

$$MD_{J_a} = \sqrt{(J_a - \mu_{M_b})\Sigma^{-1}(J_a - \mu_{M_b})'}, \quad (6)$$

where J_a is the mean of the pixels of a surface M_a , μ_{M_b} is the mean value of the mean feature of all the pixels of a different surface M_b , and Σ is the inverse covariance matrix of M_b . This

creates a new vector M_2 representing the MD of each pixel from surface M_a compared to the mean feature of a different surface M_b . This provides visualisation for the global comparison between two surfaces and semi-quantitative analysis of the degree of change (fig.4).

In conclusion, each type of data processing results in visualisation maps depicting the average measured pixel in each calculation due to the angular light reflectance. The mean feature map represents the average appearance of the surface. The mean intra-surface map provides the most prominent features that characterise the surface. Then, the inter-surface map provides a qualitative evaluation between two surfaces, a reference and a sample. In multi-temporal RTI data capture, it is essential that the reference surface remains the same and that the acquisition conditions remain constant and repeatable. In this case, the reference surface is the untarnished coupons (C0). Comparing the two surfaces using the MD does not require image registration since it provides a global evaluation; furthermore, it is possible to have a qualitative measure of the surface change with respect to the reference surface. A semi-quantitative evaluation is further feasible by extracting the mean value of the pixels calculated with the MD in the multi-temporal. This data analysis encompasses the reflectance response of all the appearance characteristics of the surface.

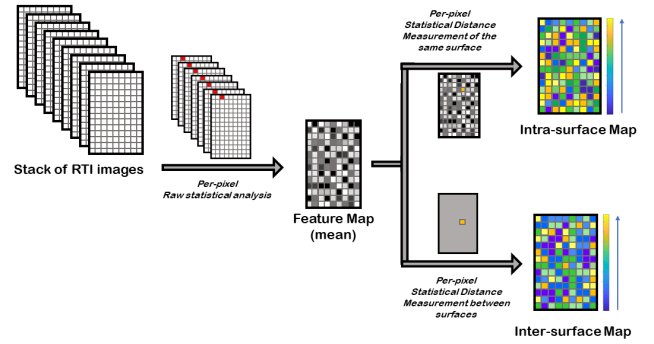


Figure 4: Flow chart of the statistical data processing for extracting features from an RTI image stack and calculating the Mahalanobis distance for inter- and intra-surface comparison.

4.3 The acquisition system

In order to ensure the resulting images' stability and repeatability, a custom-made dome with motorised functions and a single light source was used for all acquisitions. An industrial, monochromatic camera with a CMOS sensor (Sony IMX304, resolution 4112(H) × 3008 (V)) was placed at the top of the dome, and the object was placed opposite the camera in the centre of the dome. The placement was done manually since no image registration is necessary for the global evaluation of the surface; however, the same average area in the centre of the coupon was selected for data acquisition. Additionally, the same magnification was selected for all coupons and is depicted by the yellow rectangle in fig.1. A total of $N = 150$ light positions with the same homogeneous distribution were acquired for each object. The number of light positions can range from

50 to 400, and is dependant of the selected reconstruction model [37, 38]. In this case reconstruction models are not utilised, for the particular setup it is empirically known that a homogeneous distribution of around 150 positions can provide an adequate continuous angular illumination of the examined surface. Finally, due to the high specularity of the examined surface and the gradual change of the surface colour High Dynamic Range (HDR) acquisition was selected for the automatic and improved illumination of the surface [39].

4.4 Illumination Correction

The light source for RTI acquisitions must have a concentrated and intense illumination to provide enough light to the selected surfaces, even at grazing angles. Therefore, a white LED light, emitting at 6500K, with a narrow beam was used. This light provides adequate intensity; however it is not collimated, which results in a pattern with higher light intensity in the centre that decreases towards the edges (fig.6). This illumination inhomogeneity obstructs the appropriate measurement of the surface properties, which is essential to minimize its effect.

$$\mu = \frac{1}{N} \sum_{i=1}^N x_i, \rightarrow C_{i,j} = \frac{\mu}{I_{i,j}}, \quad (7)$$

where x_i being the vector containing all pixel values of the low pass filtered image (I). A correction coefficient $C^{i,j}$ is thus estimated as the ratio of the mean value μ of all pixels and each pixel value $I_{i,j}$, of the filtered image.

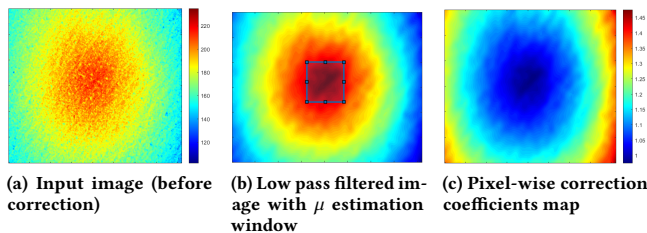


Figure 5: Flat field correction

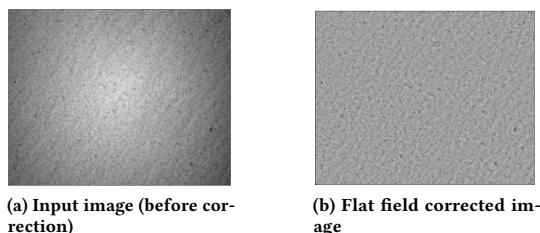


Figure 6: Flat field image correction

A flat field correction was applied by performing an RTI acquisition over a white calibration surface (Spectralon %99). The

acquisition was performed at the same angular positions and magnification used for the coupons. First, a low pass filtering was applied to the image stack to remove any interference caused by the micro-geometry of the calibration surface. Then, for each angular position, a mean pixel value of the central area (800×800 window) was estimated. Next, a pixel-wise correction coefficient was estimated for each image based on the mean pixel value of the central location as shown in equation 7. Finally, an image was obtained to visualize the resulting correction by applying the per-pixel coefficients to the raw images (input image before low pass filter). These coefficients were then used to adjust each of the acquired data sets resulting in a new corrected image set used to perform the surface analysis.

Fig. 5 provides a visualization of the light inhomogeneity in the input image (a), the selected window for the low pass filtering (b) and the areas of the application of the calculated coefficient (c). The resulting illumination corrections is presented in fig.6.

4.5 Application on silver

For the RTI acquisitions, an area of $2.4cm \times 1.8cm$ was selected in the centre of the coupon as representative of the global appearance of the surface (fig. 1). First illumination correction was applied in all data sets. Then the above mentioned methodology for the estimation of the *mean features*, *intra-surface comparison* and the *inter-surface comparison* was applied and the results are presented in figures 7, 8, 9, respectively.

Regarding the mean feature maps (fig. 7). They provide the global visualisation of the surface as a result of the per-pixel mean reflectance response for the 150 lps; the surface's general appearance can thus be visualised along with any local alterations caused by local surface variability due to the formation of the silver tarnish of the surface. However, no critical results concerning silver tarnish over time can be extracted from these visualisations.

The mean intra-surface maps (fig. 8) demonstrate the per-pixel distance of the surface concerning the mean light reflectance of the entire surface for each coupon. Here again, the surface local defects can be well visualised for the average reflectance response of the surface. Therefore the most pertinent characteristics are mapped, giving information on the relative intensity of local changes. The evaluation is done surface itself, and the scale is similar for all coupons, with a value of around 2.5. This sets the lower threshold of any measurement corresponding to non-observable change by the system.

The mean inter-surface maps calculate the per-pixel distance between a tarnished surface and the reference coupon (fig. 9), thus providing a qualitative and semi-quantitative measurement of the global surface change. In the case of the reference coupon (C0-C0), this corresponds to the intra-surface map. The most important advantage of this application is the possibility of quantifying the global change and visualising its local attributes. Areas with higher variation from the reference appear with higher values (green - yellow scale), indicating the local variation in the appearance of the tarnish layer. In terms of global change, it is noted that the highest value of the scale bar is gradually augmenting for the different time intervals depicting the gradual change in the surface reflectance due to the evolution of the tarnish.

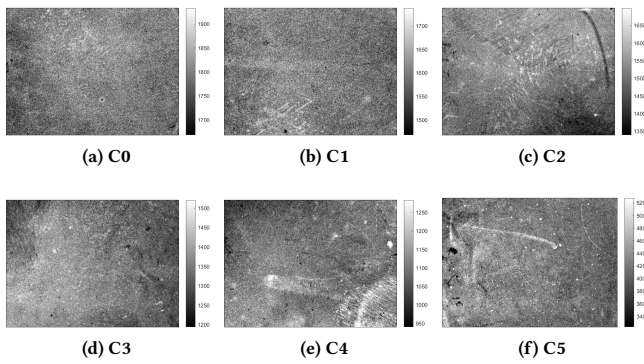


Figure 7: Mean feature map of the coupons representing different time intervals.

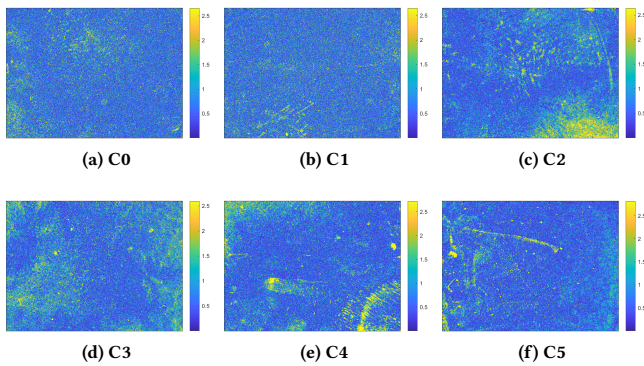


Figure 8: Calculation of the intra-surface distance of each coupon at the different time intervals.

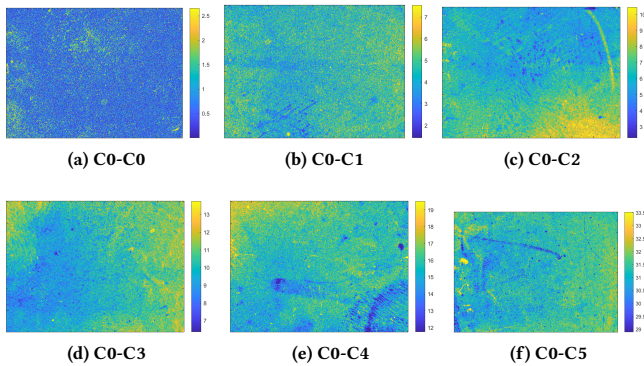


Figure 9: Calculation of the inter-surface distance of each coupon at the different time intervals in comparison to the reference C0.

In order to evaluate and quantify the information of the intra- and inter-surface visualisations, the mean pixel value of each map was calculated, and the resulting graph is presented in figure 10.

The intra-surface values for each coupon provide the baseline of the perceivable change, which is similar regardless of the degree of tarnish. On the contrary, for the inter-surface comparison, the data between the different time intervals provide a semi-quantitative evaluation of the degree of change. The resulting curve has a similar trend to the ΔE (fig. 2); though, the evaluation contains more surface information since it relates to all visual attributes of the surface calculated in a multivariate way and not only with the Euclidean distance of the colour change. In this case, the measurements are also sensitive to minor changes not perceivable by the human eye.

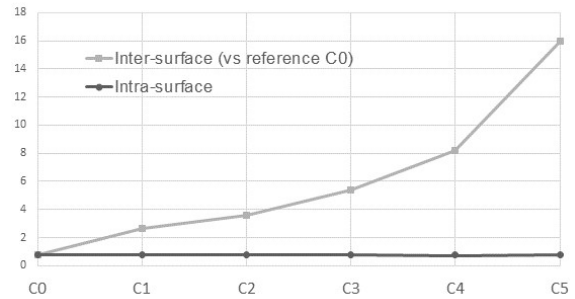


Figure 10: Graphic representation of the global mean value of the intra-surface and inter-surface measurements. Inter-surface comparison is calculated versus the reference C0.

5 DISCUSSION

The experiment was conducted with simulated surfaces on different coupons presenting similar surface characteristics but considered uniform in terms of global surface evaluation. The tarnish achieved at different time intervals is easily visible compared to the reference surface; yet, the distinction between close time intervals (i.e. C1 and C2) is not easy either by visual inspection or photography. Additionally, the edge effect highly influences human perception in assessing the resulting change in the middle of the surface. However, in a real-life situation, tarnishing creates gradual surface changes that appear on the entire observed surface, and unless induced, these changes start to become perceivable in a span of months [2]. Therefore, comparing adjacent surfaces and colour memory must also be considered. When comparing surfaces by placing them next to each other, differences are easily distinguishable in terms of colour, hue and surface texture. However, retaining the memory of visual changes of surfaces is possible only in evident differences [40].

Moreover, photography, even in controlled and calibrated conditions, cannot accurately measure the surface's change. The information registered by the RGB camera is mono-dimensional as it pertains to a single reflectance response from a diffused illumination and is highly dependent on the acquisition configuration. Thus, photography is evaluated similarly to visual inspection to document the general surface characteristic at a given instance. The resulting analysis is subjective as it depends on the viewer's perception. An appropriate methodology for measuring the surface change needs to be implemented for these purposes.

Colourimetry provides accurate results of the colour change and the lightness of the surface. The acquired results derive from a diffusive sphere in a sampling area, and multiple sampling positions are necessary for a global evaluation. This type of measurement can adequately measure only the colour attributes of the surface and not the micro-geometry. Furthermore, sampling, unless using an accurate masking process, risks a lack of repeatability and can affect global evaluation, which can be more important in the case of minimal changes. Moreover, measurements require physical contact with the surface, which is not always feasible in the case of cultural heritage objects, or may cause local alterations on the surface of soft metals like silver.

All the methods mentioned above examine mainly the colour change of the surface; RTI provides a more holistic approach as it examines the different appearance attributes of the surface as a function of their angular reflectance. Therefore, both chromatic and geometric attributes are evaluated simultaneously. Evaluating all three types of statistical analysis provides an accurate representation of the surface's global alterations while providing information on local inhomogeneities. Feature mean maps give the general surface visualization, intra-surface comparison allows mapping the salient characteristics of the surface objectively, and inter-surface comparison facilitates the qualitative and semi-quantitative evaluation of the surface change. In parallel, it is possible to visualize where a change is happening. Furthermore, one advantage of the application is that it does not require physical contact with the surface, and the sampling area can be user-defined. However, as in every MLIC method, flat surfaces are preferred. Additionally, for a global surface evaluation, it is essential that the selected area is considered uniform since the method is sensitive to surface micro-geometry; thus, any geometric change between acquisitions can affect the resulting measurement. For surfaces with complex geometries, this can be resolved by image registration. Finally, using monochromatic acquisitions, information that a change is happening can be estimated without providing the change's directionality (i.e. darker surface or colour shift). This can be overcome through RGB RTI.

6 CONCLUSIONS

Although necessary, visual inspection by experts and photographic documentation cannot fully capture or describe the changes that occur on an object, especially between consecutive time intervals with minimal surface changes. Colourimetry can provide an accurate evaluation of the surface change but in a monodimensional way since it only considers colour change. The combination of the information provided with RTI data processing demonstrates the potentiality of using RTI data processing as a tool for estimating and visualising the global surface changes in silver tarnishing based only on the surface reflectance response. Additionally, it is possible to visualise the areas where the changes are happening. The measured change describes more precisely the surface as it contains appearance attributes extending further than colour while providing an objective evaluation of the surface change.

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