Scientific analysis of historical paint and the implications for art history and art conservation. The case studies of naples yellow and discoloured smalt.
Dik, J.

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CHAPTER 6

THE DIGITAL RECONSTRUCTION OF A SMALT DISCOLOURED PAINTING BY HENDRICK TER BRUGGHEN


N.B. the section Colour Illustrations contains all illustrations referred to in this chapter

ABSTRACT

Smalt is one of the most common 17th century blue painting pigments. Smalt consists of cobalt-coloured potassium-enriched glass, that is ground to a fine, intensely blue powder. Being at the time a relatively cheap pigment, smalt was used as a substitute for other, more costly pigments roughly between the 15th and 17th centuries. As main disadvantage, however, smalt has shown to discolor over the years.

The paintings of the Dutch painter Hendrick Ter Bruggen show severe signs of smalt discoloration. A dramatic example of such degradation can be seen in his painting St Luke. This picture is part of the portrait series of the four Evangelists, kept by the Museum De Waag in Deventer, the Netherlands.

This study presents the digital reconstruction of the approximate original appearance of the discoloured painting. Neutron activation analysis and subsequent autoradiography were employed to obtain a quantified two-dimensional distribution image of discoloured paint. Digital color coordinates of undiscoloured smalt were deduced from reflectance spectra. Based on these examinations a digital reconstruction was made using image processing software. Our reconstruction shows the approximate original colours of the painting and gives insight into pictorial implications of paint discoloration in the work of Ter Bruggen.
DEUTSCHES RESUMÉ


INTRODUCTION

Like many 17th century artists Ter Brugghen made lavish use of a blue painting pigment known as smalt. This pigment is chemically unstable and has discoloured over time. A striking example of such discolouration of blue paint can be seen in Ter Brugghen’s painting of St. Luke (figure 6-1), from the portrait series of the Evangelists kept by the Museum De Waag in Deventer (figure 6-2a-c). The recent conservation treatment of this painting by the Amsterdam Atelier voor Restauratie en Research voor Schilderijen (ARR) offered a unique possibility to examine the discoloration phenomenon.

The discoloration of smalt in the painting by Ter Brugghen poses a problem to art historians, conservators and conservation scientists. These specialists are confronted with an altered appearance of the painting. The discoloration of smalt is highly detrimental to Ter Brugghen’s luminous and spatially illusive painting style. But what are the exact art historical implications of discoloured paint? What did the painting look like originally? What effects does the discoloration have on the pictorial aims of Ter Brugghen?
History of Smalt

The first use of smalt as painting pigment dates back to the late 15th century. Flemish painters, e.g. Dieric Bouts, are known to have used smalt in their paintings. Smalt is a synthetic pigment that is produced by adding cobaltarsenate to potassium enriched glass at temperatures of about 1200 °C. This solid blue coloured glass is then quenched and ground to a fine, blue powder. Mixed with linseed oil, or another binding medium, a beautiful deep blue paint results.

Smalt was a cheap and readily available substitute for other blue pigments, such as ultramarine and azurite. Ultramarine was extremely expensive, since its mineral source lapis lazuli had to imported from Afghanistan. The mineral azurite was a more widespread mineral and could be found in various regions in Europe. The purest forms of these minerals were found in copper mines located in Hungary. At the beginning of the 17th century these mines were occupied by the advancing Turks. The export of azurite to Western Europe practically came to a halt and prizes for azurite increased. As a consequence 17th century Western artists started to use smalt as a substitute on a large scale. Smalt remained the most common blue pigment until Prussian blue was discovered in the early 18th century.

Smalt has been mentioned in a few Italian and North European treatises on painting techniques. These historical sources do not indicate that discolouration was a contemporary problem, thereby suggesting that the phenomenon is indeed a slow and long term chemical process. However historical sources do focus on another notorious pigment property of smalt, its very low refractive index. As a consequence, the hiding power of the pigment is rather poor. The pigment had to be handled in a coarsely ground state, because too much grinding would cause the pigment to loose its colour intensity. Due to the large grain size of smalt, painters had great difficulty obtaining a homogeneous mixture of smalt and binding media. In their treatises Armenini, Van Mander and De Mayerne comment on this problem.

In modern literature, Joyce Plesters was the first to discuss the discolouration of smalt, describing the phenomenon on various paintings from London’s National Gallery. According to Plesters, the discolouration may be attributed to different factors. Firstly, the refractive indices of smalt and binding media may have converged during oxidation of the paint layer, resulting in a decreased capability of smalt particles to scatter incident light. Consequently, dried and darkened binding media may affect the paint system’s overall colour. Secondly, as smalt is a potash glass, potassium might be leached out into the paint medium surrounding individual smalt grains. A third possible explanation for smalt discolouration could be the migration of cobalt ions from the glass matrix into and subsequent interaction with the binding media.

In a recent study of discoloured smalt samples, Boon et al. found experimental evidence that supports the leaching out of potassium, Plester’s second possible explanation for smalt discolouration. No evidence of Co migration was found.
Digital Reconstruction of the Original Appearance of a Painting

In recent years several studies have been carried out aiming to reconstruct altered colours on paintings with the aid of digital image processing techniques. In an earlier study on discoloured orpiment, some of the current authors made such a reconstruction for a discoloured paint on a picture by Jacob de Wit. Recently, a similar study was performed at Britain's National Gallery on a 16th century Spanish painting attributed to Pedro Campaña.

It should be pointed out that such digital reconstructions are by no means an accurate portrayal of the original appearance of a painting. Colour depends on various factors such as admixture of other pigments, choice of binding media, transparency of underlaying paint, presence of yellowed varnish etc. Thus, a digital reconstruction should rather be seen as an approximation of the possible original appearance. Many assumptions have to be made when making a reconstruction, for instance the degree of discolouration, when the pigment material has not yet completely corroded. Another limiting factor is the change of refractive indices between original pigment, corrosion material and oxidized binding media. Such a change can sometimes lower the hiding power of a degraded surface pigment layer. Underlaying paint layers will become visible, which should be corrected for in a digital reconstruction.

The most problematical issue, however, of recent studies is the fact that reconstructions are made on the basis of spot examinations, rather than surface examinations. Commonly, in such studies a pigment is identified by sampling discoloured paint at a single spot. A digital image of the painting is then modified visually to match the approximate colour of the original, unaltered painting pigment. Thus, the pigment composition of a single spot is taken as the representative of a large surface area. In this way the distribution of paint over the surface of the painting, likely to be heterogeneous, is not taken into account. In such reconstructions, therefore, both the precise pigment distribution and exact colour remain unknown and have to be estimated visually.

Autoradiography of paintings

Autoradiography after neutron activation, which is in fact a two-dimensional neutron activation analysis, can visualise the original distribution of a certain paint over the painting. In fact, the distribution of a certain element is visualised, but often each colour of paint contains specific elements. For example, umber contains manganese, cobalt blue contains cobalt, bone black contains phosphor, vermilion contains mercury and smalt contains arsenic and cobalt. Even if the colour of one of such paints has changed locally, the elemental composition of the heavier elements is expected to have remained the same. For that reason, this research can help to reconstruct the original distribution of a colour and thus the unaltered appearance of the painting. Even deeper layers of paint can be visualized by autoradiography.
In this way, the distribution of umber, which was usually applied as the first onset to the painting, can be made visible.

Examinations in the past have shown that autoradiography after neutron activation is a powerful tool in conservation science, as it can visualize the working method of an artist. It has been performed on paintings from the Metropolitan Museum of Art in New York\textsuperscript{10} and the Gemäldegalerie of the Staatliche Museen in Berlin, Germany\textsuperscript{11}. However, the technique has not yet been applied in the study of paintings of Dutch Museums. Furthermore, the use of autoradiography in order to visualize the original distribution of a discoloured paint is a new application of the technique.

In this study we present a reconstruction technique in which both aspects, colour estimate of the undiscoloured paint as well paint distribution over the paint surface are deduced from experimental data. A two dimensional distribution image of the discoloured smalt was obtained through neutron activation analysis and subsequent autoradiography. This distribution image was then used as basis for further digital reconstructive work. The estimate of the approximate colour of undiscoloured smalt was made using reflectance spectrometry. Colour conversion software was used to convert the reflection spectra to a computerized R(ed)- G(reen)-B(lue) value. Thus, a digital colour tone was deduced from spectrometry data.

**EXPERIMENTAL**

**Examination of paint samples**

As part of the initial examination and in view of the interpretation of the autoradiographies several paint samples were taken from the painting. Examination of these samples included Polarized Light Microscopy (PLM) and Transmission Electron Microscopy / Energy Dispersive Spectroscopy (TEM/EDS). Cross-sections were prepared from layered paint flakes. Dispersion of the samples were examined in transmitted light using magnifications up to 1000X. Mounting medium for microscope slides was Aroclor meltmount ($N_D = 1.662$). Use was made of a Leitz Dialux EB 20 light microscope.

TEM specimens were prepared by grinding pigment samples in a hardened glass tube. A Philips CM30T TEM was used to collect data at a source voltage of 300kV. The specimen was inspected in bright field mode (BF), and grains were selected randomly for further investigations. The elemental composition was determined with a LINK EDS system attached to the column of the microscope.
Neutron Activation

The principle of the technique is that, after thermal neutron activation of the painting, a small fraction of certain elements with a high cross section for neutron absorption gets activated. After activation, the created radioactive isotopes decay to the ground state under emission of gamma and/or electron radiation. Each radioactive isotope decays with a specific half-life time (the time needed to reduce the number of radioactive atoms by 50%) of some seconds up to millions of years. After more than 6.7 half-life times, only less than 1% of the original activity of the concerning nuclide is left. If a mixture of several elements (like in a painting) is activated, the main activity at different times after activation changes from one isotope to the other because of the decay. Not all elements get as much activated: some elements have a high cross section for activation, like manganese and cobalt, while other elements like lead have an extremely small cross section for neutron absorption.

Cobalt has a high cross section for activation, though it has a long half-life. For that reason, the radiation is emitted during a long time and the activity after irradiation in disintegrations per second (Bq) is much lower than for radioisotopes with a short half-life.

The radiation emitted during decay is very specific for the radioisotope. The gamma-rays, which have a high penetrability, can be measured by gamma-ray spectroscopy with a High Purity Germanium detector. This allows quantitative analysis of all activated elements that are present in the paint. However, such gamma-ray measurements give no spatial information about the emission. On the other hand, the emitted electrons or betas have a low penetrability (0.1 to 10 mm in plastic) depending on the energy. These particles can blacken a photographic film that is in contact with the source, providing an image of the distribution of the source, but it gives no information about the energy of the electrons. This “imaging” on a photographic film is called autoradiography. By performing autoradiography at different time slots after neutron irradiation, distributions of different radionuclides can be imaged. In this way, paint distribution in various underlaying, hidden or discoloured paint layers can be imaged. Since the activity decreases with time, the time needed for an autoradiography increases with time after irradiation.

If the paint layer is deep under the surface of the painting, the electrons will often be attenuated somewhat, but depending on the electron energy and the thickness of the above lying layers, the deeper paint layer will often become visible. This implies that, if an

<table>
<thead>
<tr>
<th>Radioisotope</th>
<th>Half-life time</th>
<th>Cross section x abundance (barn)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{57}$Ti</td>
<td>5.8 minutes</td>
<td>0.0093</td>
</tr>
<tr>
<td>$^{58}$Mn</td>
<td>2.6 hours</td>
<td>13.3</td>
</tr>
<tr>
<td>$^{42}$K</td>
<td>12.4 hours</td>
<td>0.098</td>
</tr>
<tr>
<td>$^{87}$Cu</td>
<td>12.7 hours</td>
<td>3.11</td>
</tr>
<tr>
<td>$^{24}$Na</td>
<td>15 hours</td>
<td>0.53</td>
</tr>
<tr>
<td>$^{70}$As</td>
<td>26.4 hours</td>
<td>4.3</td>
</tr>
<tr>
<td>$^{197}$Hg</td>
<td>64.1 hours</td>
<td>4.8</td>
</tr>
<tr>
<td>$^{32}$P</td>
<td>14.3 days</td>
<td>0.18</td>
</tr>
<tr>
<td>$^{203}$Hg</td>
<td>46.6 days</td>
<td>1.45</td>
</tr>
<tr>
<td>$^{65}$Zn</td>
<td>244 days</td>
<td>0.38</td>
</tr>
<tr>
<td>$^{59}$Co</td>
<td>5.2 years</td>
<td>37.2</td>
</tr>
</tbody>
</table>
underpainting is present in a colour that becomes activated, it can be made visible by autoradiography without removal of the above lying paint layers.\textsuperscript{12} The first layer of 17th century paintings often contains umber, which consists of manganese- and iron oxide. Because of its high cross-section and the short half-life time, manganese is extremely suitable for autoradiography. The optimum time frame for photographic film irradiation is between 1-2 hours after neutron activation till 6 hours after neutron activation. The beta-particles emitted have a high maximum energy (2.9 MeV) and at this energy these particles are known to penetrate a few mm in plastic, and they will easily traverse a few tenths of mm of paint.

Smalt is a pigment that is very suitable for autoradiography since it contains arsenic and small amounts of cobalt. One day after neutron activation, arsenic can easily be autoradiographed. Because arsenic also emits a beta particle with a high maximum energy (3 MeV), it will produce an image that reflects the amount of smalt through the entire paint layer. The arsenic signal, therefore, may represent both the layer thickness as well as smalt concentration. The cobalt is more difficult to image because of the long half-life time (5.2 years) and the low maximum energy (0.3 MeV). The photographic film has to be on the painting for several months, the longer the better. Since the low energy betas have a very short range, they will be absorbed by even thin overlying layers of paint, and the resulting image is only the distribution of smalt on the very surface of the painting. Both the arsenic and the cobalt autoradiographs together can provide a lot of information about the distribution of smalt.

Irradiation facility

At irradiation, the painting should be exposed to a homogeneous thermal neutron flux over the entire surface. For optimal autoradiography with most sensitive photographic films, the total thermal neutron flux should be at least \(2 \times 10^{12} \text{ cm}^{-2}\). Preferably, the total irradiation time should be shorter than the half-life of the element with the shortest half-life that has to be imaged (otherwise too much will be decayed before autoradiography). Most neutron beams have the limitation that their diameter is about maximum 20 cm, which is much too small for a homogeneous irradiation of a painting, especially if the painting is as large as the \textit{Luke} by Ter Brugggen (approximately 105 x 70 cm\(^2\)). At NRG in Petten, the Netherlands, the Low Flux Reactor (LFR)\textsuperscript{13} offers a unique irradiation facility on a shielding car. Over a surface of 105 x 110 cm\(^2\), 90\% of the area is exposed to a thermal neutron flux between 7 \(10^8\) and 1 \(10^9\) neutrons cm\(^{-2}\)s\(^{-1}\). Figure 6-3 shows the thermal neutron distribution over the irradiation surface on the shielding car. The fast neutron flux is maximum 3 \(10^7\) cm\(^{-2}\)s\(^{-1}\). The surface is irradiated fairly homogeneously, the variation in intensity reaching a maximum of 30\%. In the interpretation of the final autoradiography this uneven irradiation should be taken into account.
During irradiation, the shielding car is closed and the personnel is not exposed to radiation. At 10 minutes after the irradiation, the shielding car can be opened to remove the painting. In order to prevent contamination of the painting with radioactive dust from the reactor, it is wrapped in plastic foil during neutron irradiation.

Irradiation time

From tests with paint irradiations and autoradiography trials it appeared that, using the KODAK Biomax MS films, the minimum required neutron flux is about $2 \times 10^{12}$ neutrons cm$^{-2}$. Experiments in New York have shown that irradiations with $5 \times 10^{13}$ neutrons cm$^{-2}$ did not cause any damage to the paint.\textsuperscript{14} The maximum applicable neutron fluence is determined by the rest activity when the painting is returned to the museum. Radioprotection regulations require that the maximum activity of high radiotoxic nuclides is 50 kBq for release of materials that are not subject to legislation of radioactive materials.\textsuperscript{15} The rest activity of the painting at return to the museum, (about 5 months after neutron activation) is caused by radioisotopes with a long half-life, which is mainly $^{60}$Co.

Before the final irradiation, a test irradiation was performed with 2% of neutron flux scheduled for the final irradiation. By gamma-ray spectroscopy at several points of time after test irradiation, the nuclide content of the painting was determined. From the measurements the amount of cobalt on the painting could be calculated and the rest activity of cobalt after the final irradiation could be estimated. This proved that the proposed neutron flux of $2 \times 10^{12}$ cm$^{-2}$ for the final irradiation was allowed. The final neutron irradiation was performed 2 weeks after the test irradiation.

All four Evangelists were covered with a substantially yellowed varnish, hiding most of the subtle tonal gradations in the flesh tones. In addition, discoloured retouches, notably in the facial areas, severely disturbed the appearance of the Evangelists. As part of the conservation treatment, it was therefore decided to remove the varnish as well as old retouches. Removal of varnish and retouches was carried prior to the autoradiography of St. Luke, for the following reasons. Firstly, the varnish layer would act as a filter for low energy beta’s emitted by the painting after radiation, which would result in a weaker autoradiography signal. Secondly, old retouchings are known to contain various pigments, that may consist of cadmium and other elements that would lead to nuclides with long half-life time.

Autoradiography

Autoradiography has been performed with most sensitive photographic films of KODAK, the Biomax MS films (35 cm x 43 cm). These films have a double emulsion layer, one at each side. The autoradiography is performed in a light tight box. Each autoradiography requires six films to fully cover the painting. In order to enhance the probability of beta-particles to
interact with the film, the films are covered with an intensifier foil that reflects transmitted beta-particles. A wooden board on top of the intensifier ensures close contact of the films with the painting. Table 1 shows the time scheme of autoradiographies that were performed.

<table>
<thead>
<tr>
<th>Autoradiography no</th>
<th>Time after neutron irradiation</th>
<th>Autoradiography time</th>
<th>Nuclides to image</th>
<th>Result</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0h15</td>
<td>0h05</td>
<td>Co</td>
<td>Very weak</td>
</tr>
<tr>
<td>2</td>
<td>1h20</td>
<td>0h50</td>
<td>Mn</td>
<td>Weak</td>
</tr>
<tr>
<td>3</td>
<td>2h20</td>
<td>6h30</td>
<td>Mn</td>
<td>Very good</td>
</tr>
<tr>
<td>4</td>
<td>1 day</td>
<td>1 day 2h30</td>
<td>As, Na</td>
<td>Excellent</td>
</tr>
<tr>
<td>5</td>
<td>3 days 4h45</td>
<td>6 days 16h45</td>
<td>As, Hg</td>
<td>Good</td>
</tr>
<tr>
<td>6</td>
<td>10 days 2h30</td>
<td>4 months</td>
<td>Co, P</td>
<td>Weak but good</td>
</tr>
</tbody>
</table>

During the autoradiographies, gamma-ray spectroscopy measurements were performed with a portable high-resolution high purity germanium detector. The detector was positioned on top of the box, at a distance of approximately 10 cm from the painting. These measurements allowed to identify the radioisotopes that caused the blackening of the films. For example the sodium that could be measured during autoradiography 4, appeared to be distributed homogeneously over the painting, while the arsenic could only be measured in the blue mantle of Luke. Surprisingly, also trace of isotopes like $^{198}\text{Au}$, $^{152}\text{Eu}$ and $^{140}\text{La}$ were detected. Europium and lanthanum were still unknown elements at the time of painting, and they must have appeared as a by-product in one of the applied paints. It has not been possible to identify the origin of these trace elements and the amounts were too small to influence the autoradiography.

Image processing and correction

After development the autoradiography films showing the Mn, As and Co distributions were digitalized and assembled (figure 6-4, 6-5 and 6-6 respectively). Ideally, before digitalization, some technical corrections should be applied first to each individual graytone image. First of all, as mentioned above, the neutron flux at the irradiated surface was not homogeneous. Thus, in principle a correction for the irradiation pattern, as seen in figure 6-3, should be carried out. Secondly, registration of incident radiation by sensitive photographic material is subject to a non-linear saturation curve. The speed of saturation relative to the amount of radiation received is rather slow in the lower and upper part, whereas saturation occurs rapidly in the middle part.
Both corrections have not been carried out for the following reasons. The pattern of figure 6-1 is not directly visible in any of the autoradiographs. Therefore, it can be assumed that the effect of an inhomogeneous flux distribution on the final result will be of minor importance. Unfortunately, the relation between film saturation and the amount of incident radiation could not be specified by the manufacturer of the films. Therefore, a linear relation has been assumed. An obvious improvement would be to observe the saturation curve experimentally but this could not be carried out within the framework of the current project.

Spectrometry and Colour Coordinate Calculation

Reflectance spectra were taken from undiscoloured samples of smalt using a Perkin Elmer LS 50-B fluorospectrophotometer. Smalt powder was measured with an incident beam of 45°, using variable monochromators, capable of covering the 200-800 nm wavelength spectra. The acquisition spectra were calibrated using two reflection standards, a BaSO₄ white standard (93% reflection) and black velvet (3% reflection). Graph 1 shows the reflectance curve of two modern smalt samples. Smalt Kremer denotes a commercial smalt sample produced by Kremer Pigmente (Aichstetten, Germany). The second smalt, labelled Smalt UvA, was produced by the Laboratory of Crystallography (University of Amsterdam) and the Laboratory of Solid State and Materials Chemistry (Eindhoven University of Technology). Similar reflectance spectra had been found in earlier studies. Mühletahler and Thyssen presented similar microspectrophotometric transmittance curves on a partially discoloured smalt sample from Dieric Bouts The Entombment. More recently, Boon et al. found similar spectra for smalt taken from a painting by P. Aartsen. The reflectance curves shown above were converted to various digital colour coordinates, including XYZ chromaticity coordinates and RGB Tristimulus values, using the Spectra Converter software. RGB stands for a certain ratio of standardized R(ed), G(reen) and B(ue), which translates to a specific colour. Our spectra were converted to an RGB value of R5, G35 and B255 respectively. Thus, the approximate original colour of undiscoloured smalt could be deduced from its Uv-vis absorption spectra. Represented on the colour spectra, our smalt reflectance spectra translate to a slightly greenish blue, as shown in figure 6-7.
Digital colour reconstruction

Using Adobe Photoshop 6.0 image processing software all films of each set were pasted together. The software allowed to put all autoradiographies, an X-ray radiography, and a visible light image of the painting on matching layers within one image file. Thus, all autoradiographies could be compared with each other and the visible image of the painting through matching overlays.

The graytone image autoradiography of As, showing the most pronounced contrast in smalt distribution, was selected for the digital reconstructive work. The graytone image of the autoradiography was converted to a monochrome blue image with the RGB values corresponding to undiscoloured smalt. The left side of the coat was then selected and pasted onto the visible light image of the painting. The transparency option of the overlaying mask was then set to 'hue' at 100% transparency. As a result, an approximation of the original appearance of the discoloured area of smalt was obtained (Figure 6-8). However, this process also involved a number of assumptions.

The exact relation between blackening of the As-film and the original intensity of the smalt colour could not be derived from these data. Neither could maxima and minima of the graytone image be converted to unambiguously defined saturation coordinates of the smalt RGB value. Such data therefore, had to be estimated visually. A linear function was assumed between the As-signal and the saturation of the RGB value corresponding to smalt.

A third factor that influences the relation between original colour and autoradiography blackening is the cross section for neutron absorption of each element. Since arsenic has a high cross section for activation, As-isotopes are strong emitters of radiation. Like mentioned above, smalt particles throughout the entire paint layer can be expected to have contributed to the blackening of the As-autoradiograph. The optical appearance of smalt paint, however, is dominated by light reflection of the most upper part of the paint layer. For this reason, again, the As-signal cannot be taken as a 1-to-1 linear representative of the original distribution of blue colour. It was decided, however, not to correct for these effects for the same reasons mentioned above.

In addition, the following comments should be made on the significance and function of colour reconstruction. There is a fundamentally different approach to the concept of colour by the art historian and the conservator versus the scientist. The scientist defines colour as the function of wavelength and absorption of visible light. In scientific terms, therefore, colour is defined as an autonomous entity, totally independent of its surroundings. The art historical concept of colour, on the other hand, is defined along the lines of psychological perception. The colour of a certain area perceived by the human eye is mostly dependant on its surrounding. For the scientist, therefore, colour is an absolute value, while the art historian would define colour as something highly relational. This difference in definition is of importance for the digital reconstruction of the painting. Our aim was to provide insight into
the pictorial consequences of paint discolouration. Thus, the pictorial effect of paint discolouration could only be analyzed through comparison of discoloured and reconstructed parts of the painting. For this reason it was decided to carry out a partial reconstruction of the painting, in which the left side of the mantle was adapted and the right side of the painting was left untouched.

RESULTS

A cross-section taken from the light part of the Evangelist’s mantle shows the paint stratigraphy (Figure 6-9). As usual for Caravaggist paintings, Ter Brugghen started with a reddish ground on the canvas. The first sketch of the painting was made using lead white paint to heighten the lighted left side of the portrait. Traces of this lead white underpainting are visible through thin and transparent smalt parts of the proper right arm, particularly along the elbow, which is the most brightly lit area of the painting. In our cross-section this layer lies directly on top of the ground. Ter Brugghen then continued to set up the shaded parts and toned down light areas that might appear too bright. This part of the underpainting is the thin brownish layer that consists mostly of umber and bone black. Umber is a ferrous oxide, containing small amounts of manganese. The Mn distribution, therefore, visualizes the underpainting. The presence of Mn was confirmed by gamma ray spectrometry (Graph 2). This high energy resolution spectrum was taken above the mantle during exposure of the third film. From the measurement, it can be concluded that at this location, Mn causes 5 times more exposure than As. Phosphor is the main elemental component of bone black, since the pigment was produced by burning animal bones. As explained above, phosphor shows up together with Co in the last autoradiography of the picture. Finally, the top layer in the cross-section consists of smalt mixed in varying ratios with lead white. This layer has been autoradiographed with the signal from As and Co, the main significant elemental components of smalt. The presence of smalt in this layer was confirmed through analysis of a paint sample with PLM and TEM/EDS. A microscope dispersion of a sample taken from the mantle consisted of faintly blue, isotropic particles, showing conchoidal fracture lines at the edges and a refractive index substantially lower than 1.66. Examination with TEM/EDS showed the same morphological appearance of glassy particles, consisting mainly of Si, As, Co and small amounts of K and Ni.
The As and Co autoradiographies reveal the original distribution of smalt. On both films the brushwork of discoloured smalt, lost on the original, has been made visible. A close comparison between details of the original and the corresponding autoradiographs shows the distribution of smalt down to single brushstroke resolution. Good examples are Luke’s right shoulder and the left side of his mantle, as shown in figures 6-10 through 6-13. In the autoradiography individual brushstrokes can be recognized, showing starting and ending point, the brush width and traces of brush hairs.

The mantle seems to have been executed extremely rapid and fluent, making the impression of having been painted in a single session. The shape of the mantle seems to have been enlarged at least twice. At the left and right side of the mantle additional folds of cloth were added. This might also be the reason for moving the attributes in the background, the ox and the candle, further towards the upper corners of the picture, as shown by the distribution of umber (Mn) and bone black (P). In addition, the book in the hands of the Evangelist was first painted in a slightly flatter position. In the final position the book is depicted in a more upright way. The position of the Evangelist’s head and eyes seem to have followed the modifications that were made to the book.

In general, however, the portrait was painted without making any significant changes or pentimenti. The final execution of the figure of Luke in the top smalt layer seems to follow seamlessly the underpainting in umber, used to indicate major shaded parts, such as the shadows cast by the figure’s head, his arms and hands. These positions were not changed in the final paint layer. Neither could such modifications be found in the smalt paint itself. Such changes in lower parts of the smalt layer would certainly have been visible on the As autoradiography with its high beta energy, but the film did not show any evidence for this.

The autoradiographies give further insight into the working method of Terbrugghen. The As film shows that the presence of As is strictly limited to the discoloured blue area. Therefore, smalt was only used in the blue mantle of Luke. Such a limitation in using a specific pigment for a certain passage in a painting has been noted earlier. Van der Wetering concluded that 17th century painters prepared separate palletes for different parts of the painting, like in our case the mantle. Palettes were set with a number of pigments, to be used only for a certain part in the painting. It should be noted that 17th century painters prepared their own paints and, due to technical and economical considerations, a working method as described above would have been a logical consequence.

In some areas, noticeably on the proper left shoulder and around the left elbow, the paint layer was in a very bad condition (Figure 6-14). In these areas the smalt layer seems to have been lost completely. The underlaying black background seems to be exposed because of a complete mechanical abrasion of the smalt layer, probably caused during rough cleanings in the past. Nevertheless, the As and Co autoradiographies show a weak, but profound signal for Co and As, strongly indicating the presence of smalt. In addition, on the autoradiographies the brushstrokes running through this area appear as continuous lines, without any disruption that
would suggest mechanical loss of paint (Figure 6-15). In order to clarify this contradiction a cross-section was prepared from such a seemingly smalt-abraded area. This cross-section revealed the presence of a thin layer of discoloured and entirely transparent smalt (figure 6-16). Its transparency caused the underlying dark brownish paint layer of the background to show through. The same phenomenon can be expected to have take place in the proper left side of the mantle, where the dark background shows through. Thus, not mechanical abrasion, but a complete transparency of the top paint layer seems to be the cause.

The Co film provided a similar distribution of smalt as was shown by the As film. The main difference between the As and Co distributions is that As emits a high-energetic beta that has a range of many millimetres. As a result, it will hardly be attenuated by overlying paint layers. The As signal thus depicts a distribution of both the intensity and the thickness of the smalt paint layer. This can be seen in the lower left corner where, apparently, the pen on the inkwell is painted over the mantle. The betas emitted by cobalt, however, have a very low energy and they are easily absorbed by thin overlying layers (see again the pen on the inkwell). As a result, the cobalt signal depicts a distribution of the intensity of mainly the upper part of the smalt layer. Another good example of this effect can be seen in an area where three individual brushstrokes have been painted on top of each other (Figure 6-17). The As film shows a cumulative signal, composed of all three layers, whereas the Co signal shows no distinct difference between single and multiply layered brushstrokes (Figure 6-18 and 6-19).

Four and a half months after neutron irradiation, the painting was returned to the conservator’s studio. The rest activity of the painting was lower than the background radiation level caused by radiation from the universe and construction material from the building.

DISCUSSION

Art Historical Implications

When looking at the digital reconstruction one is struck by the dramatical effect of the discolouration. It is obvious that the smalt degradation has altered the appearance of the painting severely. But what are the exact consequences – in pictorial terms – for the image of the painting? How does the discolouration interfere with our ability to perceive an image from the paint surface? And even more important: What are the consequences for a contemporary 17th century look on paintings, the way the painting was meant to be seen? In order to answer these questions we will first discuss the four Evangelists in their art historical context. Attention will be given to the Caravaggist movement in Dutch art and the pictorial aims of these artists, in which colour luminosity and illusion of depth plays a crucial role.
Terbrugghen as a Dutch follower of Caravaggio

Towards the end of the 16th century the Italian artist Michelangelo Merisi da Caravaggio, called Caravaggio, created his well-known, distinct style of highly realistic, chiaroscuro painting. This innovative painting style became quickly popular among artists from Italy, Spain, France and Holland. Hendrick ter Brugghen is usually characterized as the most important Dutch follower of Caravaggio, besides Gerard van Honthorst, Dirck van Baburen and Jan van Bijlert. This group of Caravaggists, also known as the Utrecht School, was trained by Abraham Bloemaert (1564-1651). To round up their training they all visited Italy, studying classical art, the Renaissance masters and especially the most recent art of Caravaggio. The influence of the Roman paintings of Caravaggio was enormous. His choice of themes, manner of painting, lighting, colouring and drama exercised a great attraction on young painters. Despite this influence, the Utrecht painters were never slavish imitators. Their work shows the undeniable roots of Northern Netherlandish Mannerism. In the paintings of Ter Brugghen his education in the late-manieristic atelier of Abraham Bloemaert stays visible. One of the characteristics of his style is ‘horror vacui’, a strong urge to fill the empty spots in his paintings with objects like books, papers, curtains and chandeliers. This characteristic connects him with the generation of his teacher: in series of evangelists from Wtewael and the Gheyn we recognize the same feature.

The theme of the four evangelists occurs regularly within the circles of the Utrecht Mannerists and Caravaggists. Wtewael was one of the earliest to paint the evangelists separately on canvas and panel, before him this was only done in printing. Bloemaert painted them together on canvas, although from his atelier we know also separate representations. Important series before 1621 are by Wtewael, and Baburen. Matthew, just as with Ter Brugghen, shows an intimate relation between angel and evangelist and the angel’s wing works in both series as a halo. The books and tables on Baburen’s evangelists are used in the same way as done by Ter Brugghen in his Luke and John. Although Baburen’s evangelists face the viewer more directly, his figures aren’t dressed all’ antica and his background is more quiet. Within ten years after Ter Brugghen important evangelist series followed by Jan van Bijlert, Jan Lievens, Frans Hals and Lambert Jacobs.

The prominent role of Ter Brugghen’s colourful mantles seems to suggest a special function in the iconographical scheme of the portrait series. Such a hidden meaning of the discoloured blue mantle would be a relevant aspect of the digital reconstruction of the picture of Luke. However, iconographical research did not lead to any clues to a concealed, symbolic significance of the green, yellow, red and blue togas. Therefore, the coloured mantles in Ter Brugghen’s series seem to be an iconographically arbitrary selection of colours, rather than a deliberate choice of paints with an underlying, hidden meaning.
Pictorial Aims of the Carravagists

Dramatical spatial illusion and the use of luminous colours are typical for Caravaggesque paintings. A number of painterly tricks were used to maximize the suggestion of space. The most important device is an almost theatrical lighting of the subjects in a painting. The Musical Company in London is a good example of such lighting. Like in other paintings a candle is used as single light source. Sharp shadows of drapery and the flute add to the effectiveness in creating a convincing illusion of space. Strong and clearly outlined contrasts between light and shadow are another effective tool in rendering a spatial and intimate scenery. Another effective device is an exaggerated diminuendo of light intensity, as pointed out by Van de Wetering. Objects close to the light source are depicted brightly lit, sometimes making the impression of 'overexposure'. Backgrounds are usually kept much darker, even relative to the distance of the light source.

References to this pictorial effect can be found in a few art theoretical sources. Interestingly, in their treatment of light the Caravaggists seem to go against the advice given by various art theorists. Willem Goeree, for instance, wrote in his treatise that daylight from above is the ideal lighting situation, because daylight:

creates soft and unified shades and the bodies keep their shape through simple reflexions in a natural way as they show themselves without restlessness and hardness.\(^3\)

Long before Goeree another authoritative source, Leonardo da Vinci, had already warned against hard contrasts due to strong illuminations, commenting that

This device is to be employed seldom in painting, because such works are crude and without attractiveness (crude e senza grazia).\(^3\)

It almost seems as if the Caravaggists with their harsh contrasts in light and shadow and strong single light sources deliberately opposed textual descriptions as mentioned above. Crude and harsh lighting conditions were used intentionally in order to maximize spatial illusion. In addition, Caravaggio and his followers categorically abstained from using soft, blurred or 'smokey' contour lines, described by the Italians as *sfumato*. According to contemporary sources this device, again, was a tool to give a painting 'sweetness' or 'softness'. The Caravaggists instead, are known to have used strong, sharp edges between bordering paint areas. As Van der Wetering suggested, crudeness or hardness, as opposed to 'attractiveness or sweetness', might just have been the deliberate pictorial aim of Caravagist painters.
Another typical feature of Caravaggesque painting is the use of highly luminous colours. Strong hues of red, yellow and green can be seen in most pictures by Terbrugghen. It should be noted that such colours are hardly subordinated to the lighting situation of the scene depicted. Colour retains its autonomy, as described by Van der Wetering. This is exemplified by the fact that white highlights are given a slight, but noticeable hue of the underlaying colour. For instance, red parts of a costume are given reddish highlights, each part having his own highlight. Such ‘pedantic’ treatment of colour cannot be found with other painters of the 17th century. The optical result, however, is the experience of independent and strongly luminous colours in the painting.

Pictorial effects of discolouration

The use of brilliant colours and convincing suggestion of spatial depth, therefore, plays a prominent role in Ter Brugghen’s oeuvre. Both pictorial aspects, however, are severely disturbed by the discolouration of smalt. First of all, the discoloration affects the suggestion of space. This becomes clear when comparing the reconstruction with the discoloured original. In figure 6-8 the left side of the coat has been reconstructed, while the right side has been left unchanged. The left side conveys the sensation of depth and three-dimensionality to the viewer, while the right side of the coat remains a greyish flat area without any suggestion of space. Any contrast, or even marks of brushstrokes are no longer visible in this part of the picture. The folds on the left side of the coat and the shadows cast, however, help the eye to understand the three-dimensional relations in this part of the picture. In addition, the transparency of the smalt layer has caused underlaying paint layers to shine through, as can be seen on Luke’s proper right side, where the lead white underpainting has been exposed. This not only interferes with the suggestion of depth in the picture, it also has disastrous consequences for the imitation of the texture of the mantle. The pigment has become transparent, which has caused the underlaying paint to shine through the smalt layer. The mantle no longer appears as a texturized, spatial drapery, but rather as flat, uncomprehensible layer of paint. Thus, loss of suggestion of depth is the most evident consequence of paint discolouration.

The second effect can only be evaluated when comparing the discoloured Luke with his fellow-Evangelists. Matthew, John and Marc have been preserved in excellent physical condition and have not suffered from discolouration phenomena. As mentioned above all Evangelists are depicted in a similar setting, wearing bright, colourful togas. It should be noted that the colours used, blue and yellow and red and green respectively, are so called ‘constrasting colours’ which are perceived by the human eye as distinctly opposite. It is quite conceivable, therefore, that the hue of the prominent colourful togas of the Evangelists must have been carefully balanced in order to produce a harmonizing series of portraits. As a
consequence, the degradation of smalt disturbs the colour harmony in these portraits. Or, in less subjective words, the ensemble of colours in the portraits has been changed.

In a 21st century perception, both aspects of spatial illusion and colour harmony are separate characteristics of the visual arts. No necessary connection between both issues is felt when looking at paintings. Colour harmony for instance can be enjoyed in an abstract painting without feeling the urge to undergo the illusion of spatial depth.

It should be stressed that this separate experience of colour harmony on one side and spatial illusion on the other was not shared by the 17th century viewer. On the contrary, in the 17th century perception colour harmony and illusion of space were seen as strongly intertwined aspects of painting. This combination of colour harmony and spatial illusion has been referred to as “houding” in contemporary 17th century theoretical sources. The word ‘houding’ in modern Dutch is translated as ‘attitude’ in English. Paul Taylor, however, has elucidated the complex double meaning of the concept of Houding in 17th century artistic vocabulary.36 The leading Dutch art critic of the Dutch Golden Art, Samuel van Hoogstraten gives the following description of the concept of ‘houding’

Thus it should be commented on metaphorical order and beautiful arrangement. This secret of art we are common to express by ‘Houding’. And in colouring this means the same as symmetry, analogy, harmony and proportion in the art of arrangement. It is also similar to conformity and a charming way of singing in music. Because it involves a pure gathering of harmonizing force: the arrangement of colour, which we call the ‘Tuylkonst’. And the righteous arrangement of lights and shadows as well as pulling forward, pushing backwards, rounding and foreshortening. And it finally leaves nothing excluded of the things mentioned above which are part of a perfect painting.37

This paragraph stresses the double concept of houding, which combines harmonic ideas of colour with a convincing suggestion of space. This latter part of the concept of houding is expatiated on in another contemporary theoretical work by Willem Goeree. Goeree writes:

‘Houding’ is one of the most essential things one can observe in a drawing or painting, because it gives us the same optical experience that we enjoy when looking at things in nature. If ‘Houding’ cannot be found in an artificial image, such drawings or paintings have no reason and appears are more than half dead. Through the lack of ‘Houding’ things appear mixed up, packed together tumbling against us. In such a way that nothing can be more against reason than to place things without ‘Houding’. It is necessary to show what ‘Houding’ is and through which actions it can be obtained. Houding—in order to express the meaning of this art term and the force of its essence—is that which connects everything in a drawing or painting, which makes
recede or advance and puts everything in its place, without making it appear closer of
further away, nor to make it lighter or darker than its closeness or distance allows. In
a way that everything, without confusion, loose from others in its neighbourhood,
stand on its own place, and keeps its size and colour, light and shadow. So that one
can detect with one’s eyes the space in between, the distance between each object,
receding or advancing, as if it were accessible by foot and firmly planted on its place:
and this one calls ‘Houding’.

The 17th century viewer of paintings, therefore, wanted to undergo illusion of space combined
with an aesthetical experience of colour harmony. As described above the discolouration of
smalt destroys colour balance and illusory depth. Much worse for the 17th century eye,
however, is the disastrous effect on the subtle and highly esteemed relation between both
aspects. The unity between colour and spatial deception is completely ruined. In 17th century
words the houding of the portrait of Luke is lost. The modern viewer is able to tone down the
effects of discolouration by experiencing colour and spatial depth as separate entities. As
demonstrated above the 17th century viewer had a much more integrating look on paintings,
which did not allow for a separate and selective experience of both aspects. In the 17th century
perception of painting therefore, the effect of discolouration must have had truly disastrous
consequences. Much more so than the present day museum visitor realizes.

CONCLUSION

In the study above we described the digital reconstruction of a smalt discoloured painting,
Luke the Evangelist, by Hendrick ter Brugghen. Such digital reconstructions have been
carried out in the past, but in this article a new methodology is introduced. Our method for the
reconstruction relies on surface rather than spot examination, providing local experimental
data on original colour. It was shown how a combination of autoradiography, colour
digitalization of undiscoloured smalt and image processing resulted in an image of the
approximate original appearance of the painting.

Comparison between the present day painting and the reconstruction allowed us to evaluate
the pictorial consequences of paint discolouration. It was shown how the discolouration
would have been experienced by a 17th century viewer of the painting. Given the importance
of the notion of houding in the 17th century art theory it was concluded that the discolouration
is much more disturbing for the 17th century perception of painting than for the present day
witness.

We feel that a digital reconstruction can be a powerful tool in raising awareness for
sometimes dramatical material changes in works of art that occasionally remain unnoticed by
art historians. In addition, we hope to have demonstrated that such material changes go hand
in hand with optical and pictorial alterations that have direct consequences for art historical interpretations.

Autoradiography after neutron irradiation proved to be a valuable method to learn about the set-up of a painting. The distribution of different paints could be visualised with sub-millimetre resolution, showing precisely every brush stroke that the painter had originally made. Even small changes that the painter made in the set-up could be visualized. Especially smalt is an interesting colour for autoradiography because it contains two elements, As and Co, which can be visualized individually without interference of other elements.

SPECIAL REMARK

A documentary about this project was made by VPRO television, Hilversum, the Netherlands. ‘De Mantel van Lucas’ can be ordered as VHS video or can be viewed as a streaming video on the website of VPRO television at http://www.noorderlicht.vpro.nl

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NOTES


2 J. A. De Graaf, Het De Mayerne Manuscript als bron voor de schildertechnieken van de Barok, Mijdrecht, 1958, p.149

3 G. G. Armenini, De veri precetti della pittura, Ravenna, 1586

4 C. van Mander, Den grondt der edel vry schilder-const, fotomechanische herdruk, Utrecht, 1973, leerdicht XII, 43

5 J. A. De Graaf, 1958, p.42-43


8 M. den Leeuw and J. Dik, "Door de Konst getovert voor myn oogen' : Over de schildertechniek van Jacob de Wit", in: In de Wolken: Jacob de Wit als plafondschilder, Bijbels Museum, Amsterdam, 2000, p.72-86


12 ibid.

13 The LFR is a small reactor of the Argonaut-type with a maximum thermal power of 30 kW.

14 See note 10, M. W. Aisnworth 1982, chapter 3

15 as prescribed in Dutch nuclear safety legislation (Besluit Stralenbescherming Kernenergiewet). As a comparison, human beings contain an average of 100 Bq/kg bodyweight.

16 Another radioisotope, $^{55}$Fe with a half life of 2.7 years, emits such low energetic radiation that it cannot be measured.

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17 details on the reconstruction of smalt to published elsewhere

18 see note 1, p. 116

19 see note 7, p. 957

20 by the Dept. of Information Technology of Lappeenranta University of Technology, Lappeenranta, Finland. During our research in the fall of 2001 the Spectrum Converter software could be accessed through the internet at http://www.it.lut.fi/research/color/lutcs_database.html

21 The use of red ochre ground is characteristic for Caravaggist painters, see M. Koller, “Das Staffeleibild der Neuzeit”, in : H. Kühn, Farbmittel, Buchmalerei, Tafel- und Leinwandmalerei, Stuttgart, 1984, p. 350


23 the pictorial consequence of discoloured paint was discussed earlier by the authors in Den Leeuw and Dik, 2000, see note 8


27 from one series, to be dated 1610-1615, two paintings are in the Rijksmuseum, Amsterdam. A second series, dated 1616, is in Stuttgart

28 The series of Baburen in La Seo, Saragozza is the most important

29 one in Utrecht, one in Belfast

30 all four in Bamberg

31 two in Odessa, one in the Paul Getty Museum

32 all four in Rouen


34 W. Goeree, Inleydinge tot de algemene Teyken-konst, Middelburg, 1670, p.63-68, cited from op. cit. Van de Wetering 1988, note 8
Zoo resteert er noch van der zelver overeendrachtige ordening en meedevoeglijke schikkinge te spreekten. Welke geheymenis der konst wy gewoon zijn met het woort Houding uit de drukken; 't welk in het koloreeren even het zelve beteykent, als konst der maatschiklijkheyt der woorden Simmetrie, Analogie, Harmonie, en Proportie; zijnde ook als de overeenstemming en bekoorlijke zangwijze inde Muziek. Want het begrijpt in zich een zuivere vergaderinge van samenstemmende kracht : het wel schikken der koreuten, 't welk wy de Tuilkonst noemen; en de ordenlijcke schikking van lichten en schaduwen; nevens het voorkomen, wechwijken, ronden, en verkorten; en laet eyndelijk niets uitgeslooten van al't gene dat, booven 't geene dat reets verhandelt is, tot een volmaekte Schildercke behoort.

De Houdinge is een van de nootwendigste dingen die in een Tekening of Schilderye moet waargenomen worden; om dat zy ons het zelve gevoel aan het oog doet krijgen, dat wy in' t beschouwen van de natuurlijke dingen genieten. Want wanneer de Houding in de nagebootste beeldhuiwe niet gevonden word, zijn sulke Teikeningen en Schilderyen redenloos, en meer dan half dood. Ook schijnen de dingen, door de ontbeering der Houding, alle malkanderen verward, op een gepakt, of tommelings tegen ons aan te vallen; suks dat er in de geheele konst niets gedaan kan worden, dat meer tegen de reden aan kan loopen, dan dingen sonder Houding te plaatsen. 't Is noodig, dat wy toonen wat de Houding is, en door wat opmerkende daad men die moet sien te bekomen. Houdinge, om den zin van het konst-woord, en de kragt van hare natuur uyt te drukken, is dat gene, welk alles in een Teikening of Schilderye verbond word, doet agter en voor uyt wijken, en alles van het voorste tot het middelste, en van daar tot het agterste, op sijn eigen plaats doet staan, sonder nader of verder te schijnen, noch figer of donkerder te vertoonen, als sijn ver-heid of nabyheid toelaat; invoegen yeder ding, sonder verwering, los en wel uyt andere die 'er nevens en omtrent zijn, en op sijn eigen stant-plaats, soo wel van grootte als van kleur, ligt en schaduwe gehouden blijft; ja dat men de tussen-ruimte van de plaats, of distantie die tussen yder lichaam open en ledig is, van zig wijkende, of na zig toekomende natuurlijk met het oog, als of het met de voeten toegankelijk ware, kan naspeuren, en op sijn plaats geplant vinden: en dit noemt men Houdinge.