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PAINTINGS

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ABSTRACT

Recent macro-XRF scanning of Rembrandt's *Self-portrait* from 1669 in the Mauritshuis – as part of the ReVisRembrandt project – has revealed significant new information about the pigments and build-up of the painting. The elemental distribution maps make clear that the umber-rich ground plays a very important role in the final appearance of this painting, information that is highly relevant for the study of this self-portrait, as well as many other late Rembrandt paintings. It was also possible to image the presence of organic lakes (by means of the potassium map) and bone black (by means of the calcium and phosphorus maps) in the upper paint layers. In addition to conventional investigative techniques, such as x-radiography

INTRODUCTION

Distinguishing between past restoration, the effects of ageing and deliberate painterly effects can be complex, and often provides challenges for paintings conservators. This is particularly the case with Rembrandt's late paintings. The informal technique, rough texture and dark palette of these works, in combination with surface degradation problems and layers of overpaint, can make them difficult to interpret. Traditional imaging techniques, x-radiography and infrared photography/reflectography often provide little information about the paint layers. For this reason a multi-interdisciplinary, international research project ReVisRembrandt (2012–17) was started, as part of the Science4Arts initiative co-funded by the Netherlands Organization for Scientific Research (NWO) and the American National Science Foundation (NSF).[1] During the ReVisRembrandt project, conservators, scientists and art historians are working closely together to develop and apply new imaging techniques to the study of late Rembrandt paintings. Techniques include x-ray fluorescence (XRF) imaging (also referred to as macro-XRF scanning), hyperspectral and multispectral infrared imaging spectroscopy and x-ray diffraction imaging. Complementary to these new imaging methods, existing paint cross sections of late Rembrandt paintings are being studied with a range of laboratory- and synchrotron-based micro-analytical techniques, in order to provide a full elemental, structural and molecular characterization. When combined, these techniques will: a) identify a wide range of painting materials and degradation products and b) plot their distribution over the surface of a painting and its substructure. For late Rembrandt paintings, this will provide a deeper understanding of the sequence of paint layers, artist changes and condition in order to answer open questions regarding original appearance, painting technique and attribution. An important goal of the ReVisRembrandt project is to link Rembrandt's pictorial goals to technical features in his late paintings, namely his materials and working methods.

MACRO-XRF SCANNING OF LATE REMBRANDT PAINTINGS

Selected late Rembrandt paintings – both informal as well as commissioned portraits and history paintings – are being investigated with mobile XRF scanners developed at the University of Antwerp and by Bruker Nano GmbH (Alfeld et al. 2011 and 2013, Noble et al. 2012). This novel macro-scanning

and infrared imaging, it is anticipated macro-XRF scanning will become an important diagnostic tool for conservators and art historians alike.

technique makes it possible to visualize the distribution of elements in both surface and subsurface layers of a painting in a non-invasive manner at a resolution down to 0.1 mm. Separate black and white distribution images are produced for each element in the painting where light areas represent areas in which a high intensity of the element's fluorescence radiation was recorded. The maps can then be used to make interpretations about the pigment composition, painting technique and condition.

In 2012, investigation of Rembrandt's *Self-portrait* from 1669 (Mauritshuis, The Hague) took place using the first commercially available, mobile XRF scanner (Bruker *M6 Jetstream*).[2] The main body of this paper presents the results of that investigation. Since the scans contain contributions from all paint layers, it is not always straightforward to determine if an element is present in a surface layer or in an underlying layer. Moreover, surface layers can absorb part of the fluorescence radiation emitted by subsurface layers; elements with lower energetic radiation (such as phosphorous or potassium) especially can be completely blocked by overlying paint layers. In order to facilitate interpretation, XRF scans were correlated with paint sample analysis and examination of the paint surface with the stereo-microscope.[3] The painting was also investigated with IR photography and comparison was made to an existing x-radiograph of the painting in order to make a full comparison of all the analytical data. As part of the ReVisRembrandt project, visible and near-infrared imaging spectroscopy of this painting also took place in March 2014.

REMBRANDT *SELF-PORTRAIT,* 1669

Figure 1
Rembrandt, *Self-portrait*, canvas, 65.4 x 60.2 cm, signed and dated 1669, Royal Picture Gallery Mauritshuis, The Hague

Rembrandt's late *Self-portrait*, which is signed and dated 1669, is a fine example of the expressive freedom that characterizes Rembrandt's late works (Figure 1). It is considered to be the last of three self-portraits (Florence, London and The Hague) painted in 1669, the last year of his life. Treated in the Mauritshuis in 2005, the painting is in excellent condition, and as such is considered to provide a touchstone for Rembrandt's late painting technique. The portrait is painted on a linen support, on top of a dark grey-brown ground. The paint layers on the whole are thinly applied with the exception of the face, which is built up with vigorously textured passages containing lead white. The multicoloured headdress is also applied in multiple layers as a result of having undergone revision. Investigation in the past with x-radiography had revealed an initial white painter's cap, a change Rembrandt made during the painting process (Figure 2, left image), although it is not known to what extent it was worked up before being modified. In keeping with the portrait's informal character, the abbreviated handling leaves the brownish ground or undermodelling visible in small areas around the eyes, mouth and neck bordering the white collar. At some stage in the painting process, an additional background paint, slightly lighter in colour, was added around the head. The description of the painting technique published in *A Corpus of Rembrandt Paintings IV* in 2005, largely based on a report supplied by the Mauritshuis in 1998, before the painting was treated, makes mention of a brown sketch or undermodelling (Van de Wetering et al. 2005, 588–593). Part of the goal of the recent investigation was to

Figure 2
Left: X-radiograph. Right: XRF distribution map of lead (Pb-L)

Figure 3
Left: XRF distribution map of iron (Fe-K). Right: XRF distribution map of manganese (Mn-K)

come to a better understanding of Rembrandt's late painting technique and try to image the preparatory layer(s). For the study of Rembrandt paintings in particular and 17th-century paintings in general, this feature – when it does not contain carbon-based black – is of particular interest as it is not visible on the surface of the painting and up until now could only be made visible by neutron activation autoradiography (NAAR), an established method for visualizing the elemental composition of a painting. However, since this technique requires transport to and a stay of several months at a research reactor, it is infrequently applied.

MACRO-XRF SCANNING RESULTS AND DISCUSSION

In this painting, the lead (Pb-L), manganese (Mn-K) and iron (Fe-K) maps are the most informative about the stages in the build-up of the painting. In Figure 2 (right image) the Pb-L map is shown, the light areas representing areas of high intensity. Like the x-radiograph shown at the left, it clearly demonstrates the lead (white) in the ground layer that is in close contact with the canvas. The Pb-L map also shows the initial white painter's cap visible in the x-radiograph. It is striking that this earlier, smaller cap, hidden below the thick paint layers, shows up more clearly in the x-radiograph as compared with the Pb-L map. This is primarily because the x-ray fluorescence radiation, which is of lower energy (10–12 keV), as compared to the much higher energy of the primary beam employed in traditional x-ray radiography, is more strongly absorbed in the covering paint layers. Not visible in the x-radiograph, but noticeable in the Pb-L map, is the light halo around the head. Comparison of paint cross sections (not shown) confirmed that an additional application of lighter grey-brown paint containing more lead white was added around the head before the revision to the headdress was made.

For Rembrandt paintings, it is usually assumed that Rembrandt made a sketch or undermodelling (so-called *doodverf*) in brown, black or muted tonalities that were subsequently worked up. The manganese (Mn-K) and iron (Fe-K) maps (Figure 3) might at first suggest the visualization of a preparatory sketch with umber, a brown earth pigment. However, when examining the areas of the painting that demonstrate a high Mn signal anew under the stereo-microscope (a large part of the background and shadow areas in the face and neck), it became clear that it is the grey-brown ground that is exposed in these areas. This was subsequently confirmed by paint cross sections from the background and neck, which revealed a single layer corresponding to the ground (Figure 4). The composition of the ground was recently identified as comprising umber, lead white, yellow and red ochre and fine charcoal black.[4] With this information in mind, the Mn-K and Fe-K distribution maps can be interpreted as the umber and ochre in the ground, where the light areas correlate with areas of the ground that were left exposed and the dark areas where the signal is blocked by overlying paint layers. This becomes even clearer when comparing the manganese and the lead distribution maps. The light areas in the Pb-L map, where lead-rich paint is present, corresponds to exactly the same areas that appear dark in the Mn-K distribution map (Figures 2–3). Thus, it is the ground that is exposed in small areas around the eyes, mouth and neck and not an undermodelling layer, which would be expected to have a different composition than the ground. Thin areas of exposed ground (slightly abraded on the tops of the canvas threads) are also noticeable in a detail of the infrared photograph of the face and neck (Figure 5). That Rembrandt left the ground exposed to form a large part of the background represents new information about the painting.[5]

As already described in previous publications, Rembrandt changed the initial painter's cap into a fanciful multi-coloured headdress. The distribution maps make it possible to characterize the nature of the paint and to visualize the extent of the revision. Examination of a paint cross section from the hat along with the iron (Fe-K) and potassium (K-K) maps demonstrates that one or two red paint layers rich in fine bright red earth and yellow lake were used to define the higher turban-like hat (A *Corpus of Rembrandt paintings* IV erroneously refers to a layer of red lead). On top of the red layer, the headdress was then finished wet on wet with broad light-brown stripes/brushstrokes containing large amounts of red and yellow lake pigments, in addition to yellow and red earth and bone black. Accordingly these strokes appear light in the Fe-K (Figure 3, left image) and K-K distribution maps (Figure 6, top image).

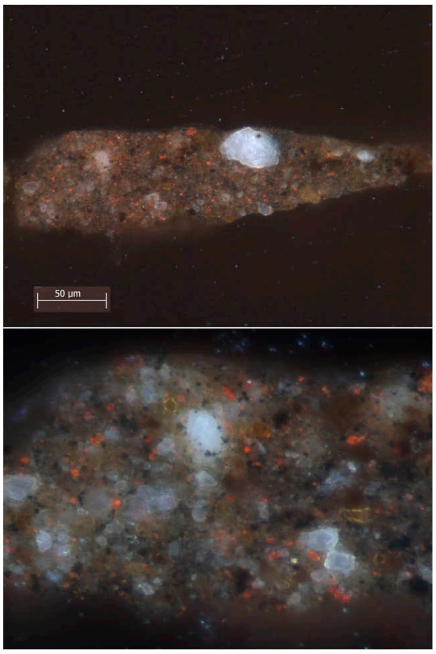

Figure 4
Paint cross section of exposed dark grey-brown ground comprising umber, lead white, yellow and red ochre and fine charcoal black in shadow area of the neck (Mh840x24). Top: normal light, dark field, photographed at 400x magnification. Bottom: normal light, oil immersion, photographed at 1000x magnification

Figure 5
Detail of digital infrared photograph showing the thin ground exposed in shadow areas in the face and neck (*Artist* camera, IR2: 900–1100 nm)

The potassium (K-K) distribution map also shows that lake pigments were used exclusively in the flesh tones to impart a pink tonality (Figure 6). In this portrait, mercury, associated with the pigment vermilion (HgS), was not detected in the flesh paints, nor in the reddish areas of the headdress (Hg-L map not shown). The relatively thick application of lead white in the face, which is more readily visible in the lead map (Figure 2) – as compared to the x-radiograph – was subsequently worked up with numerous touches of dull red-brown earth, especially in the corner of the mouth, below the earlobe and on the nose.

One of the intriguing features of the painting is the presence of blackish borders preserved on three sides (originally on four sides) that would have functioned as a sort of preliminary framing. Paint sample analysis revealed that the borders were painted directly on top of the ground with a mixture of bone black, organic brown (possibly Kassel earth), yellow and red lake, as well as a little smalt and lead white. That the paint indeed contains lake pigments was established by EDX micro-analyses of paint cross sections (EDX: Al, Ca, S, K, Cl). It is notable that the potassium (K-K) distribution images appear to be a remarkably good marker for the lake pigments (Figure 6), the potassium possibly the result of alkaline extraction of the dyestuff, whereby the excess potassium salt was not completely removed (Kirby et al. 2005, 77). This is consistent with the relatively high amounts of potassium identified in the lake substrates with EDX spot analysis.

Traditionally, infrared imaging is used to image carbon black in paintings. For late Rembrandt paintings, however, this is hampered by the absorption of infrared when black is present in both surface and subsurface paint layers, as is the case with this self-portrait and many other late Rembrandt paintings. Successful imaging depends on the thickness of the paint layers, wavelength and sufficient contrast with underlying layers that reflects infrared, such as a white ground. Since in this painting bone black was used for the black garment, and is located on the surface of the painting, the distribution of this pigment could be successfully imaged with macro-XRF scanning by means of the phosphorous (P-K) and calcium (Ca-K) distribution maps (Figure 7).[6] The broad brushwork of the bone-black mantel is dramatically captured in the calcium map, including what appears to be the use of the finger in the wet paint to create a detail of the cloak. The paint cross sections reveal that the cloak – in this sample, a mixture of bone black with a little red and yellow lake – was applied

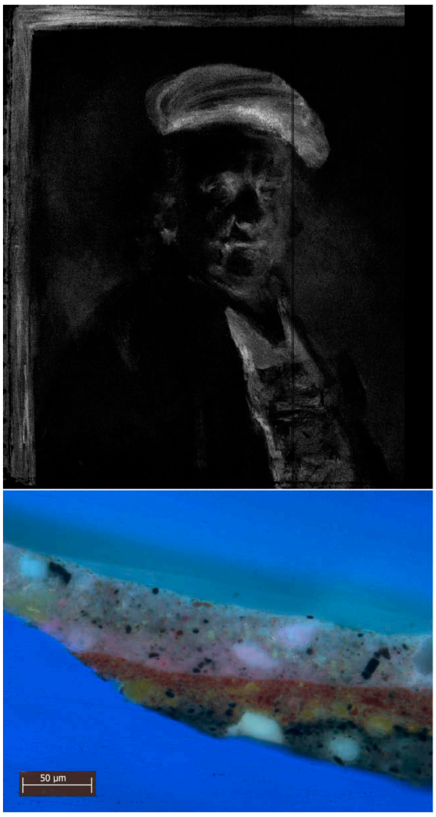

Figure 6
Top: XRF distribution map of potassium (K-K). Bottom: paint cross section (MH840x21) from brown stripe in the headdress. This layer appears strongly fluorescent due to the presence of lake pigment particles (ultraviolet light, photographed at 400x magnification)

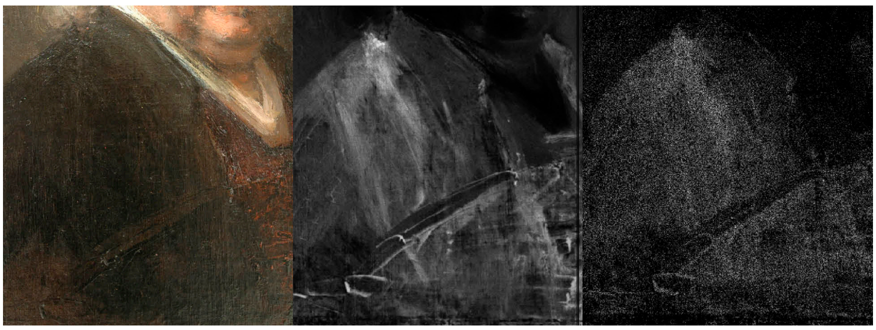

Figure 7
Left: Detail of cloak in visible light. Centre: XRF distribution map of calcium (Ca-K). Right: XRF distribution map of phosphorous (P-K)

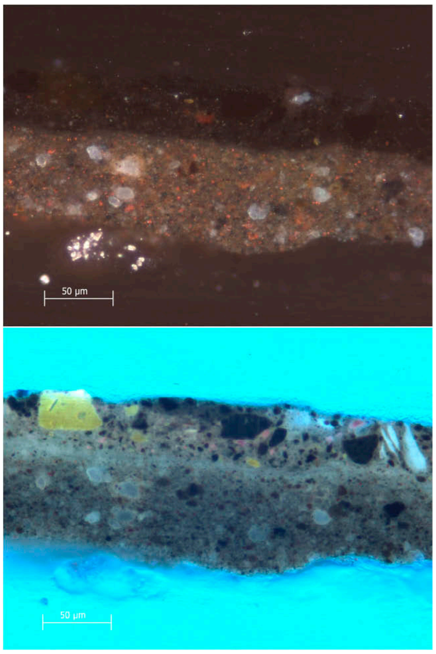

Figure 8
Paint cross section of black mantle (Mh840x20) showing two black layers on top of the grey-brown ground. Top: normal light, dark field, photographed at 400x magnification. Bottom: ultraviolet light, photographed at 400x magnification

in either one or two layers over the grey-brown ground (Figure 8). It is important to realize that since bone black, made of charred bones, and characterized by the presence of $Ca_3(PO_4)_2$ in addition to carbon, contains light atomic weight elements, P and Ca, it is not possible to image this pigment when located deep in the paint structure, for instance when a bone-black sketch is suspected. In that case it would be more appropriate to use neutron activation autoradiography (NAAR).[7]

Interestingly for this self-portrait, painted in the last months of Rembrandt's life, there appears to be no brown or black sketch. Instead the artist appears to have made expedient use of the brown ground. By painting directly on a dark ground, he did away with the traditional sketch/undermodelling, allowing the ground to serve as the background and shadow areas in the painting. This short-cut method may explain the more provisionary, sketchy nature of the portrait as described by Van de Wetering (Van de Wetering et al. 2005, 593). Indeed, comparison with the *Self-portrait* from the National Gallery London, also dated 1669 and considered to be painted only a matter of months before the The Hague picture, reveals a more complex build-up and sophisticated palette.[8] This shows that even though the two paintings are of the same subject and painted only some months apart, there can be considerable technical differences.

CONCLUSIONS

The recent macro-XRF scanning of Rembrandt's late *Self-portrait* – as part of the ReVisRembrandt project – has revealed significant new information about the distribution and identification of pigments, as well as the build-up of the painting. The elemental distribution images make clear that the umber-rich ground plays a very important role in the final appearance of this painting, information that is highly relevant for the study of this self-portrait, as well as the many other paintings by the late Rembrandt.

Compared to traditional x-ray radiographs, the absence of the stretcher bars in the XRF distribution maps facilitates a clearer reading, although lead-white features located deep in the paint structure may show up more strongly in the traditional x-ray radiographs as compared to the lead (Pb-L) distribution map, but this will largely depend on the nature and thickness of the overlying paint.

As mentioned, interpretation of the macro-XRF data can be complex and prior knowledge of the painting technique and expected pigments/elements is crucial. Correlation with information from paint cross sections is desirable.

The development of new imaging techniques opens up new avenues for the non-invasive study of artists' materials and methods, making it possible to study the substructure and condition of paintings in far greater detail and within a reasonable time frame. For the late Rembrandt in particular, the creative process can be studied much more accurately. It is also anticipated that macro-XRF scanning will provide significant information for resolving questions regarding workshop variations, attribution, dating, authenticity and the state of preservation of these enigmatic late paintings.

ACKNOWLEDGEMENTS

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NOTES

1 Partners include Delft University of Technology (main applicant) and the Royal Picture Gallery Mauritshuis (co-applicant), together with the University of Antwerp, National Gallery of Art (Washington), Synchrotron Soleil (France), Rijksmuseum Amsterdam, The Netherlands Institute for Art History (RKD) and the Wallraf-Richartz-Museum (Cologne, Germany).

2 Scanning parameters: Rh-tube, 50 kV, 600 µA, 750 µm step size, 100 ms dwell time per pixel, map size: 635 mm × 565 mm (847 × 752 pixels). Depending on the elements under investigation, the step size and dwell time should be carefully determined, but this also depends on the time allocated for scanning of the painting.

3 All paint cross sections were examined in the Mauritshuis using the light microscope (Leica DM2500). SEM/EDX analyses were carried out on an XL30 SFEG high-vacuum electron microscope (FEI, Eindhoven, The Netherlands) with an EDAX system for elemental analyses at FOM Institute AMOLF, Amsterdam, 2013.

4 In contrast to earlier research by Groen (2005, 670–671), bone black was not identified in the ground layer, but rather fine charcoal black; the large dark particles in the grounds were identified as manganese oxide-rich umber (SEM/EDX). The presence of finely distributed yellow ochre in the ground was made visible with a 1000× oil immersion lens in the light microscope.

5 Before the conservation treatment of the painting in 2005, the background was partially covered in overpaint.

6 The phosphorous (P-K) distribution map was initially considered to be due to an escape peak for calcium that falls nearly exactly on the P-K line. After careful comparison, it became clear the phosphorous map truly demonstrates the presence of phosphorous associated with bone black in the painting.

7 Recent comparison of the imaging capabilities of macro-XRF scanning, as compared to neutron activation autoradiography (NAAR), demonstrated the advantages and disadvantages, as well as the differences between the two methods. The elemental distribution maps acquired by macro-XRF scanning are easier to interpret as all elements are clearly separated. However, with NAAR, the resolution of the images is considerably better, but this depends on the macro-XRF scanning parameters (Alfeld et al. in preparation).

8 Bomford et al. 2006, cat. no. 21, 190–195. Figure 212 shows brown undermodelling below the flesh paint. Painted on a quartz ground, this painting has a different, more complex build-up.

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