# SOULAGES' *PEINTURE 12 MARCH 1960*: DIAGNOSTIC ANALYTICAL STUDIES AND TENTATIVE CONSOLIDATION OF MAJOR CURLED-UP DELAMINATED AREAS

## Abstract

The paper describes one of the most extreme forms of delamination seen in works by Pierre Soulages. It concerns a painting titled with its date of creation *Peinture, 162 x 200 cm, 12 Mars 1960* presently in the Sarah Hildén Museum in Tampere Finland. The delaminating surface paint has formed open wounds with curled up paint revealing an interface with the commercial ground where loss of cohesion started. Simultaneously, the upper bone black paint layer is forming acidic oxidized oil-derived exudates that also penetrate the lower paint layers. Imaging microscopic and spectroscopic methods reveal details of a dynamic process of delamination with paint consistency changes and mixing of layers near the delaminating interface during loss of cohesion. First successful experimental consolidation tests to reverse the open wounds were successfully carried out.

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Pierre Soulages; delaminating paint; exudation; loss of cohesion; medium acidification; consolidation.

## Introduction

Pierre Soulages (b.1919), still active after eight decades of work and the most renowned French painter alive today, is known for his exclusive use of the colour black in non-figurative works. He began his career with black oil paint strokes that contrasted and overlapped with lighter grey, yellow ochre, red or blue paint. Those paintings played an important role in the development of a new form of abstraction based on marks, lines and tracks of painting tools in the 1950s. Soulages developed large tools, used broad brushes, and adapted his oil media to render his tool traces visible. Unfortunately, a small number of oil paintings made in 1959 and 1960 are showing signs of severe damage in the form of fissures, cracks, and delamination, with extreme lifting of the paint layers. The cause for the appearance of these disturbing phenomena is thought to be the reactivity of underlying oil paint layers with the ground. These phenomena are particularly prominent in a painting in the collection of Sara Hildén in Tampere, Finland (Fig.1). The difference between the condition of *Peinture 12 mars 1960* recorded previously and its disastrous present state provided the rationale for a detailed multispectroscopic and microscopic study. The affected areas were compared with those in good condition. Raman spectroscopy, imaging Fourier Transform Infrared (FTIR), Scanning Electron Microscopy (SEM) and Gas Chromatography -Mass spectrometry (GC-MS) were applied together to identify and determine the distribution of organic and inorganic materials in selected cross-sections and samples relevant to the understanding of the problem of the delamination. The study presented here is performed to provide a scientific examination of the observed phenomena and to assist in the development of a conservation strategy. Initial conservation experiments to explore the counteracting of the lifting of paint layers are reported.

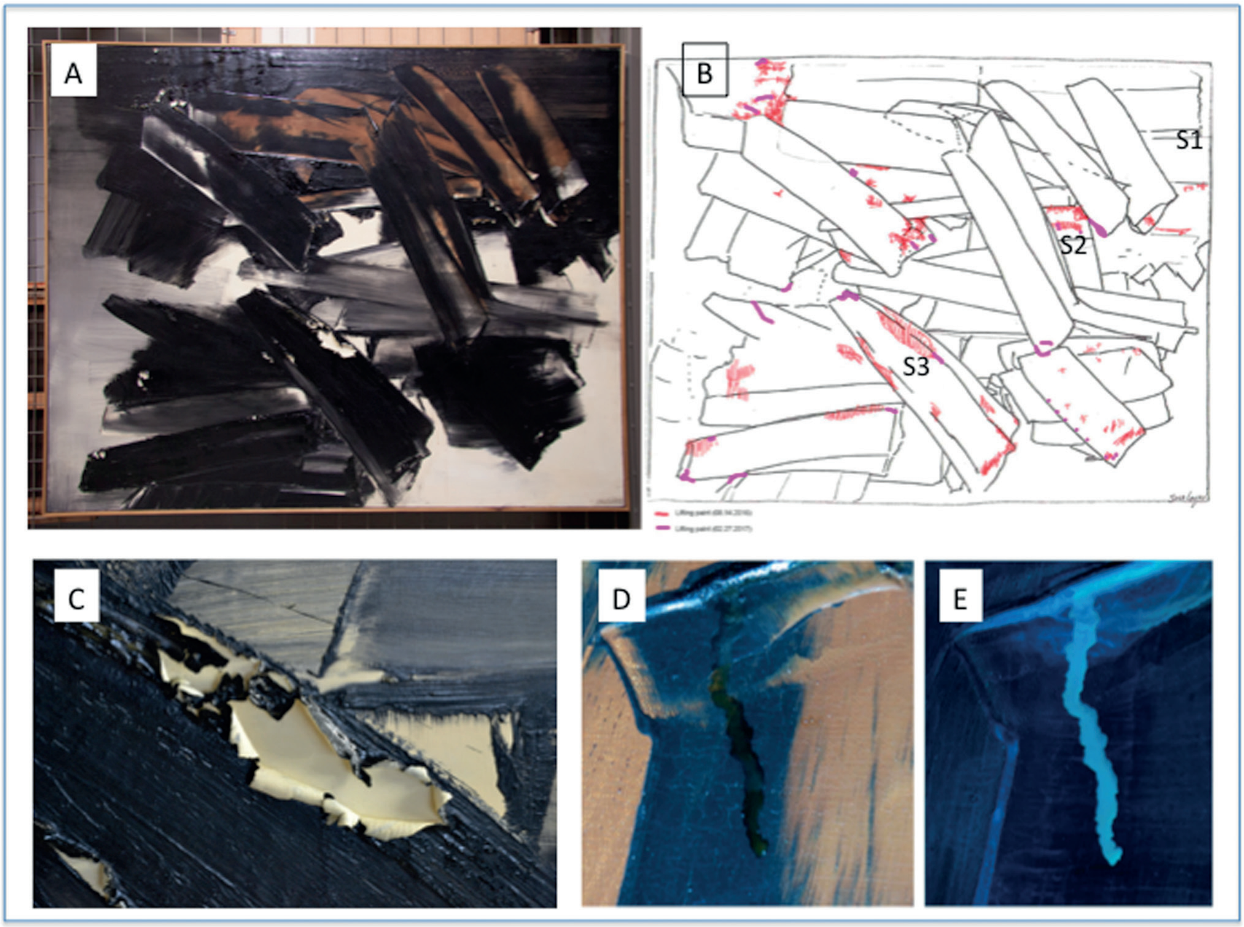

Figure 1. Areas of lifted paint on *Peinture 12 mars 1960* by Pierre Soulages (162 x200 cm, oil on canvas, Sarah Hildén Museum, Tampere, Finland). Photo of the painting (A). Drawing of the main paint strokes and areas of damage with numbers indicating the sample positions (B). Major delamination area with curled up paint (C). Impasto and exudate drip in daylight (D) and fluorescent light (365 nm) (E). Photos: Hélou-de La Grandière and Boon.

## Degradation phenomena of some Soulages paintings

The delamination phenomena have been seen in several of Soulages' paintings from 1959 and 1960. In his painting at the Musée d'Art moderne de Paris, *16 Décembre 1959*, the lead white priming was suspected to be linked to cleavage defects (Hélou-de La Grandière et al. 2008). In this painting and some others dated around Decembre 1959, the commercial ground from colourman Lefebvre-Foinet with plumbonacrite and lead soaps was thought to be the cause of the delamination. Paintings by other artists who used the pre-primed canvases from this paint supplier also suffered various related defects (Hélou-de La Grandière et al. 2008, Hélou-de La Grandière et al. 2009, Corbeil et al. 2011; Bronken and Boon 2014; O'Malley 2019).

In addition to the delamination phenomena, the surface layer in some paintings from around 1959-1960 have shown dripping exudates. A Soulages painting titled *Peinture, 130 x 162 cm, 17 mars 1960* in the Art Institute of Chicago has exhibited this defect. Specifically, an acidic fraction that developed from oxidizing oil paint in the bone black paint appeared at the surface as dripping exudates (Boon and Lister, 2014). The same weeping degradation had also been observed in a contemporary work of Soulages (*Peinture 15 Décembre 1959*) from Toulouse, France (Hélou-de La Grandière 2017).

Also possibly linked to the colourman Lefebvre-Foinet, compositionally similar exudates have been found in paintings by Jean-Paul Riopelle (Bronken and Boon 2014) from the 1950s and in paintings by Karel Appel from that time, during his Parisian period (Bronken et al. 2019 ; Boeve-Jones et al. 2020). The damage concerns not only the change in surface condition of the works, but also inherently affects the paints, leading to softening of the oil paint internally, and the formation of new materials. Additionally, it causes internal weakening affecting the structure of impastos leading to loss of cohesion where paint layers tear and are lifting. These issues have been described as aspects of the "Montparnasse Disease" (Hélou-de La Grandière 2019).

Such defects developed over time in the paint layers, so it is difficult to determine cause and effect: Has painting technique played a role? Are the issues related to the drying conditions? Or environmental conditions? These questions are currently being studied in a PhD thesis project by the first author entitled "NOIRŒS"[1]. So far, each new case study is adding to the understanding of the degradation process, such as the painting from Tampere presented in this paper.

All of the earlier case studies were done during conservation treatments. They highlight the importance of the constitutive elements, with interactions between the industrial ground and the paints (industrial or mixed by Pierre Soulages himself). Although the focus is on paintings with conservation issues, not all paintings by Soulages from this period feature such signs of damage. There are as yet unknown factors involved.

## The painting process of Pierre Soulages

To fully understand Pierre Soulages' painting process, the first author had the opportunity to ask the artist about his methods since first meeting him in 2004. Priorly, Roger Vailland (1907-1965), a poet and friend of Soulages, was present in his studio during the creation of *Peinture, 200x160 cm, 27 Mars 1961*, and wrote a paper "How Pierre Soulages works?" (Vailland 1961). Because this painting is quite similar in composition, format, and was made not long after our case study Peinture, 12 mars 1960, the making process may have been very similar.

Vailland describes the scene, summarized as follows: In his large and sunny Parisian studio, Soulages takes time to prepare the materials and canvas surface before proceeding to paint. He feels that the ground is too greasy, so he first degreases it with a cloth and "an appropriate liquid"[2]. However, this degreasing goes too far, so, after a dozen minutes, he applies a kind of jelly with a brush to make the canvas more slippery again. Vailland then describes the preparation of all the colours by Soulages: *"he places a tank filled with ivory black paint, which he finds to be too thick and therefore thins by adding "an oily liquid". In another bin he empties a large tube of white oil paint*"[3]. Then, he mixes the colours on transparent glass. The colours are placed, scraped off and replaced on the canvas over the course of three hours, using brushes and a rubber slab.

This eyewitness account highlights how Pierre Soulages paid attention to the quality of the ground even when commercially manufactured. It also shows that he created his own melange using commercial paint tubes and additives to obtain the desired consistency. Of course, sixty years later and after more than a thousand paintings, Pierre Soulages does not recall the exact recipe of this "appropriate liquid", the "jelly" nor the additives in the preparation of the paints. However, he remembers which materials from Lefebvre-Foinet he used - three different types of commercial pre-primed canvasses (available with variable porosities). He also recalls that he chose the "fatty preparations", because he wanted to avoid the excessive absorption of binders and to avoid a matte appearance to the paint. According to his recollection, this type of preparation consisted of three coats of lead white ([Soulages 2004], de La Grandière 2005, Hélou-de La Grandière 2019). The contemporary suppliers confirmed that each was applied as soon as the previous one had dried on the surface (de La Grandière 2005:58).

This priming process from the colourmen suggests short drying times between each layer, a procedure that can affect the lead white pigment (Leback-Stiwell 1989; Zucker 1999). In France, the lead white ban in 1948 led to a number of manufacturers going out of business. Thereafter the supply of the lead white became problematic and the quality of canvas preparations very variable, as was confirmed by the colourmen working in Paris at that time (Edouard Adam, Dominique Sennelier, see de La Grandière 2005). Soulages has himself blamed Lefebvre-Foinet:

> *« At the end of the fifties, I had, as well as some comrades who had the same supplier as me, yet very famous, some canvases whose priming layer dissociated itself from the support[4]»* (Soulages, 1994: 54).

## Present condition of the painting

*Peinture 12 mars 1960* was bought soon after completion by the Finnish businesswoman and art collector Sara Hildén (1905–1993) and presently resides in the Sara Hildén museum in Tampere. The painting has been kept in storage for years. Figures 1A and 1B illustrate the extent of the damaged areas on this painting. Figure 1B is a map of the damage observed on the painting with sampling areas marked as S1, S2 and S3. Lifting and severe delamination of the paint layers, deformation, softening of the layers, accompanied with dripping of medium rich exudations and changes in surface quality (excessive gloss) appear everywhere where black surface paint is present, except where the red-ochre-brown paint is the undercoat, as well as in thinly-painted parts. The delaminated area that is about 15cm in length in Figure 1C shows surface black paint that is acutely curled up, revealing the off-white underpaint beneath. In general, the upper black layer suffers from a strong modification of its characteristics as wave-shaped deformations are visible and correspond to a softening of the paint. These deformations were observed to evolve rapidly when the painting was moved from storage to the museum space for examination - possibly due to a sudden change in environment. Softening seems linked to the lifting process and is also coupled with a strong increase in the glossiness of the surface, which emits a strong bluish fluorescence under UV light (365 nm). Ultimately, the softening either leads to a sticky dust-attracting surface or to exudation of an UV fluorescent fluid (Fig. 1E).

## Paint build-up and spectroscopic studies

Samples could be taken from several delaminated areas (S2 and S3), the exudate and an undisturbed black painted area near the right tacking margin (S1). Sample areas were photographed in situ. 3D-Hirox-microscopy was performed in the laboratory before further microsampling, embedding in Technovit 4000, UV-curing for 20 minutes and subsequent hand-polishing of the blocks with a series of micromesh polishing clothes (1800 – 8000 mesh) to obtain cross sections suitable for analytical studies. The cross sections were investigated at the Art Technical Laboratory in Bern with visible light, UV light, Raman spectroscopy  and FTIR microscopy before investigation by SEM under electron back scatter conditions (BSE) and Energy Dispersive X-ray element mapping (EDX). Subsamples were also taken for investigation by GC-MS using the on-line analytical transmethylation protocol (Van Keulen 2014). For detailed experimental conditions see section *Equipment.*

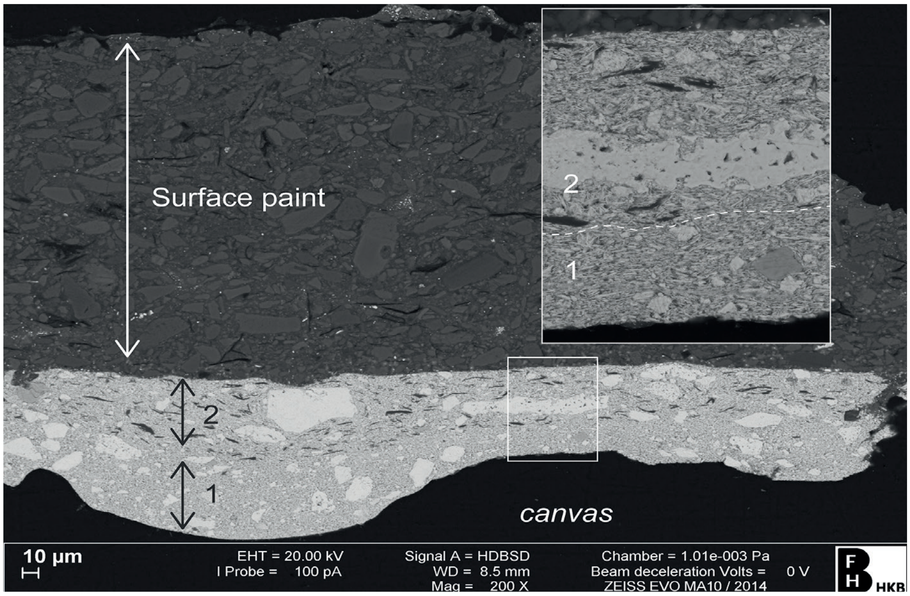

Figure 2. SEM-BSE image of the paint layer buildup near the right-hand side tacking margin (S1 in Fig 1B) showing a double ground with layer 1 with large fragments of lead white in a finer matrix and layer 2 with lead white and elongated particles of talc (dark in SEM-BSE). Layer 1 follows the canvas surface. A thick layer of bone black paint forms the top layer. Photos: Scherrer & Boon.

All cross sections were studied under visible and UV light. However, the layer structure and particle composition are best understood through Scanning Electron Microscopy imagery, particularly electron backscatter images (SEM-BSE). The cross section of the sample at the tacking margin (S1), shown in Figure 2, illustrates the ground and a simple black paint layer, which overflowed from the composition. Figure 2 is composed of two parts: an overview and an inserted magnified area of the ground layers. The priming is a double-layered ground consisting of a lower layer (1) 30-40 microns thick, of fine crystalline lead white mixed with large lead white chunks and occasionally a rounded particle of barite (grey under BSE). The second, similar lead white layer (2) above shows many elongated hydrated magnesium silicate particles of talc (black under BSE). A thick layer of black paint (120 micron) with particles of bone black (calcium and phosphorous detected with EDX) is present. The painted area at the tacking margin is solid and shows no sign of weeping or loss of adhesion. This part of the painting will be compared with those areas where the delamination occurred.

The SEM-BSE image in Figure 3 of the cross section of the second microsample (S2) taken from a slightly lifting paint area shows a more complete set of paint layers. Layer 1 is broken off at the bottom on the right-hand side upon sampling. Layer 2 is marked by the elongated talc particles. The first layer applied by Soulages (3) is a finely-divided, very well-sorted lead white layer corresponding to the off-white underpaint visible on the surface of the painting below the whole composition (Fig. 1A). This is followed by the black paint, divided in two layers in our examination: layer 4, considered to be a transition zone with a mix of lead white from layer 3 and bone black particles suggesting a wet-in-wet technique, and then layer 5, composed of black paint alone. The image taken under UV light shows that layer 3 has a slightly different fluorescence hue compared to the other layers. This layer (3) shows the FTIR characteristics of aliphatic chains at 2953-2889 cm$^{-1}$(vC-H), esters at 1765-1722 cm$^{-1}$(vC=O), lead soaps at 1565- 1504 cm$^{-1}$(vC=O) and lead white carbonate at 1455-1360 cm$^{-1}$. The top layer has a peculiar blue fluorescent and transparent interlayer between layers 4 and 5 (denoted with a star). This layer has a strong unique absorption in the FTIR range from 1724-1660, pointing to ketones and acids upon FTIR mapping. This could be an entrapped exudate.

The stratigraphy of this cross section is remarkable. The top layer (5) is curled up together with the layer (4) underneath. The particles in layer 3 in the middle of the section are oriented like the ones from layers

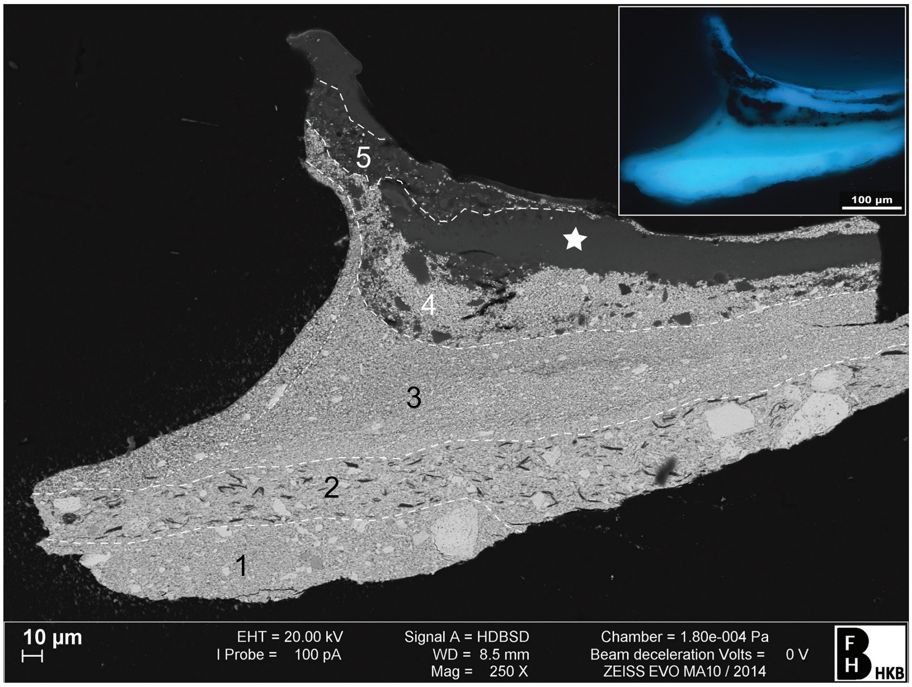

Figure 3. SEM-BSE collated image of the paint layer buildup in the center of the painting (S2 in Fig 1B). Layer 1 and 2 form the double ground. Layer 3 is the off-white underpaint used to create the background. Layer 4 is a wet-in-wet layer of 3 and 5. Layer 5 is the black bone black surface paint. The insert is a microscopic photo under UV light. The star is a carbon rich inter layer. Photos: Scherrer & Boon.

1 and 2, but are almost perpendicularly oriented on the left side (dashed line left) where it became exposed to the open air during delamination. These changes in stratigraphy suggest that layer 3 must have undergone a change in consistency while the area was lifting.

A third sample (S3) investigated was a paint flake that had broken off in the major delamination zone shown in Figure 1C. Microsamples were taken from this paint flake for examination with imaging-light microscopy, imaging FTIR-microscopy and SEM-EDX, to determine the paint layer build-up and composition. Raman spectroscopy was employed for the identification of specific mineral phases.

Microscopic examination with 3D-Hirox microscopy showed the off-white underside as a corrugated surface covered with a film of light-reflecting "wet-looking" exudates. The surface of this off-white paint exhibits distinctive ridges (Fig. 4), which are known as cohesion fillets in adhesion science (SpecialChem 2021; Pocius and Dillard 2002). The pattern of these ridges suggests that the underside had at some point become softened, with changed viscoelastic properties leading to a loss of cohesion and to detachment from the remaining ground below. Certainly, the curling of the lifted paint borders indicates that the delaminating paint stack was ductile during the separation process. A similar corrugated surface was present in another sample from a different zone of lifted paint. Light reflected from this surface also suggested the presence of a thin coating of light-reflecting organic materials.

The SEM-BSE picture of the cross section of the paint flake S3 (prepared through one of the ridges) shows the loss of a clear layer structure (Fig. 5). The top paint layer (5) and the layer below (4) are still discernible but layer 3 and below appear to have become an unexpected mixture of layers 2 and 3 (indicated as 2+3). The dark flat talc crystals of hydrous magnesium silicate present in the second ground layer (2), and identified with Raman spectroscopy, are suggestive markers for an increasing fluidity that had affected the second ground layer and the lower part of Soulages' off-white underpaint. The bottom of the cross-section (the interface where the delamination has occurred) shows depletion in solids.

The cross section was investigated with SEM-EDX element mapping and Attenuated Total Reflection (ATR)-FTIR-microscopy. The footprint of the ATR crystal of the FTIR microscope on the cross section is shown as a white-lined box in the SEM-BSE image

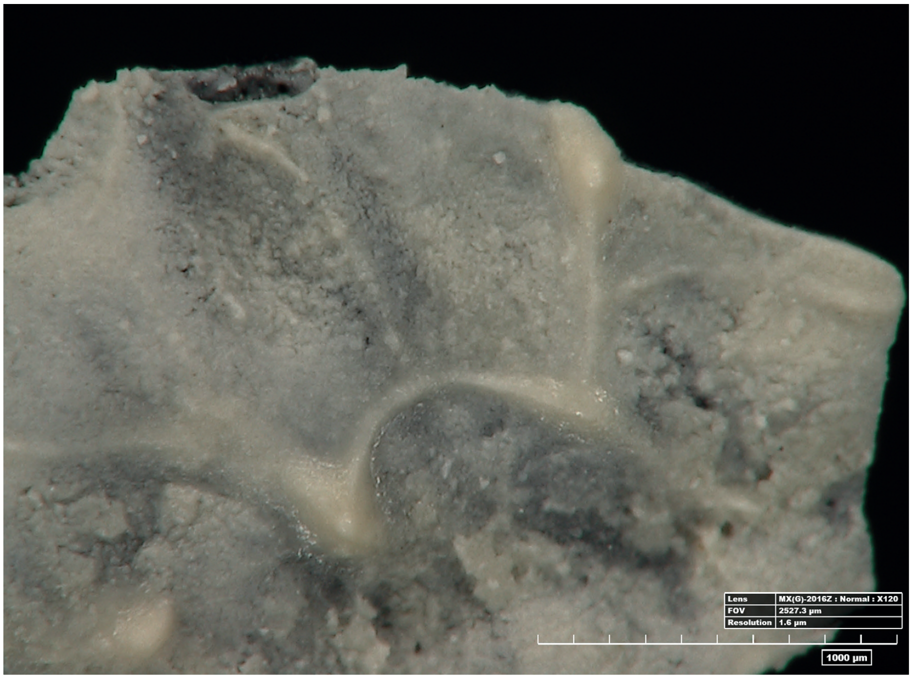

Figure 4. Hirox 3D-image of the underside of a paint flake sampled from the major area of delamination (S3 in Fig 1 A) showing ridges that suggest plastic deformation during delamination from lower layers. Photo: Boon.

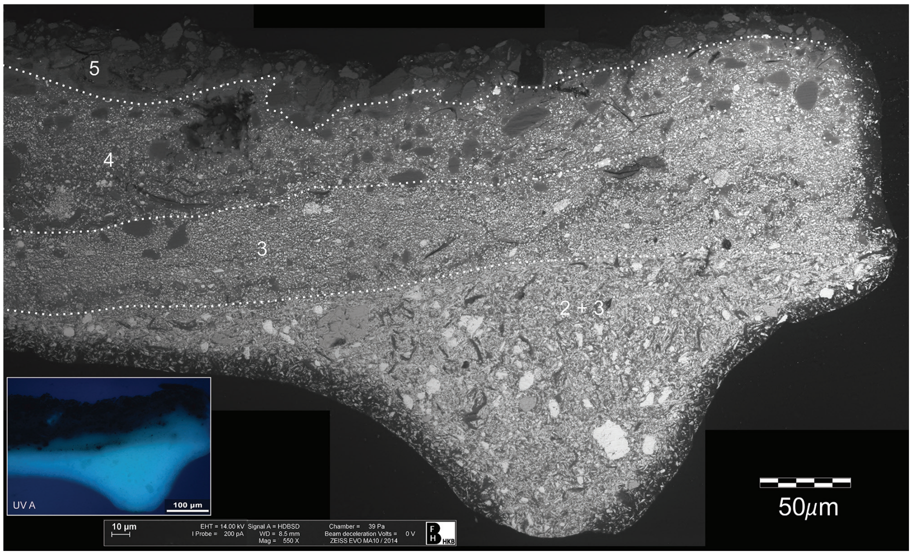

Figure 5. SEM-BSE image of a cross section through a ridge in the delamination zone. The numbers correlate with the layer buildup in Fig. 3. The layer 2+3 is a mixture of layers 2 and 3. Photo: Scherrer & Boon.

in Figure 6. This area was chosen for FTIR imaging because it includes part of layer 4, layer 3, the mixed layer (2+3) and the thin delamination zone (D).

Figure 6 combines the maps from FTIR-Focal Plane Array imaging and SEM-EDX. The SEM-EDX imaging maps show the distribution of lead (Pb) in purple, and mineral phase maps[5] of magnesium silicate (talc), calcium phosphate (bone black) and barium sulphate (baryte). The talc is the marker of the second ground. The maps represent the relative signal intensity of the spectral windows denoted above each map.

The FTIR maps of carbonates at 1464-1334 cm$^{-1}$(Fig. 6), the map of general binder features at 2949-2898 cm$^{-1}$ (asymmetric CH2 vibrations) and esters at 1761-1725 cm$^{-1}$ (vC=O) correlate well with the lead distribution (Pb M-lines), pointing to lead carbonates and lead soaps. The FTIR map of saturated fatty acid soaps (lead stearates) 1545-1493cm$^{-1}$ correlates well with the off-white underpaint in the top part of the stratigraphy. The lower part of the stratigraphy with the talc (layer 2+3) shows some differences in distribution of (lead)-carboxylates (1576-1517 cm$^{-1}$) and acids (1735-1691 cm$^{-1}$). This difference in lead soap distribution with respect to the off-white paint could point to an alternative structure of the lead in the lead soap of layer 2+3 (Hermans 2017). The increased acidity at the bottom could be linked to the identified acidic molecules observed with GC-MS (see below). The ~10-micron-thin delamination zone represents an area of strongly-reduced pigment and binder content (see fig. 6). Indeed, this boundary layer seems highly weakened.

Figure 7 illustrates a SEM-BSE image at high magnification of the boundary layer of paint flake S3 with a thickness of 10-20 microns. The elongated crystals within the delamination zone are interpreted as cerussite. The FTIR images in Figure 7 show the distribution of binding medium features (1759-1685 cm$^{-1}$: C=O from esters), the distribution of carbonate (1459-1325 cm$^{-1}$) and hydroxyl groups (3564-3491 cm$^{-1}$) respectively. The boundary layer is depleted in hydroxyl group features. It has a higher concentration of cerussite relative to the layer above with hydrocerussite and plumbonacrite both identified with Raman spectroscopy (785 nm). Hydrocerussite and plumbonacrite are stable under alkaline conditions but cerussite [$PbCO_3$] is more stable under weaker alkaline conditions with a boundary in the phase diagram at about pH 8.5 (Taylor and Lopata 1984). The distribution of these lead mineral phases suggests that the cohesion loss, leading to weakening of the interface to ground layer 1 and the process of delamination, could be the result of pH changes. Studies with micro-X-ray diffraction would be desirable for further confirmation of the changing mineral phases.

## Binding medium analysis

The molecular signature of the delaminated paint layer and the exudate were investigated by GC-MS on-line analytical transmethylation protocol

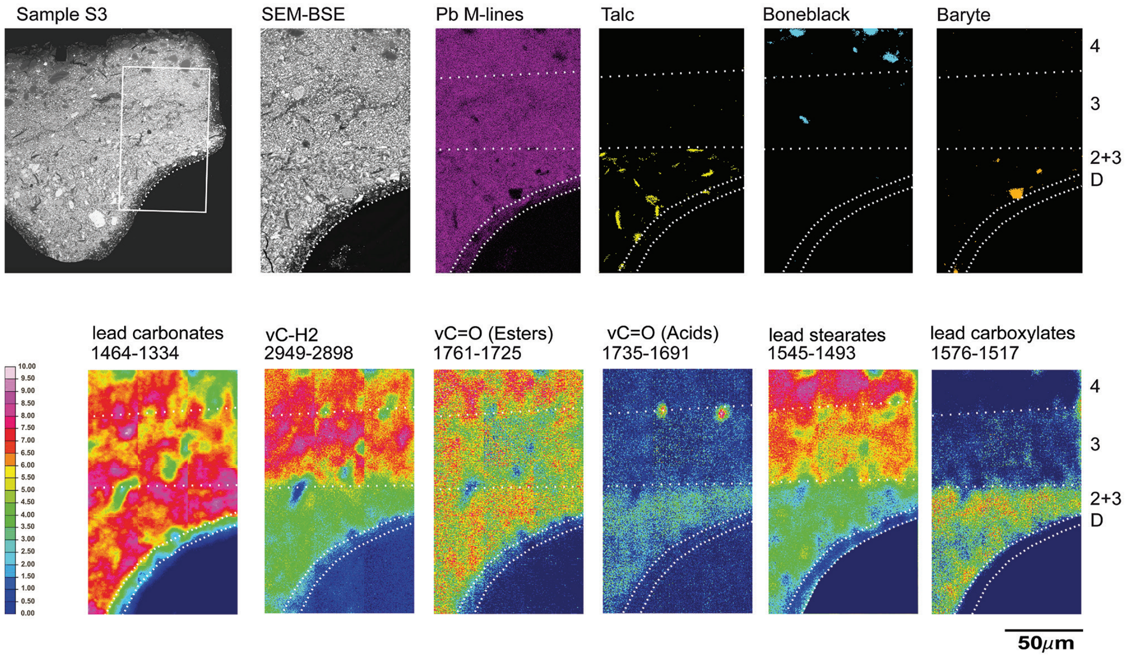

Figure 6. SEM-BSE overview and detail of sample S3 with SEM-EDX element and phase maps, and FTIR-FPA images of the right-hand side of the cross section in Fig 5. FTIR-FPA images represent the relative intensity of the corresponding spectral window denoted above each map. Dashed lines and associated numbers on the right-hand side indicate the various paint layers and the delamination zone (D). Illustrations: Zumbühl & Scherrer.

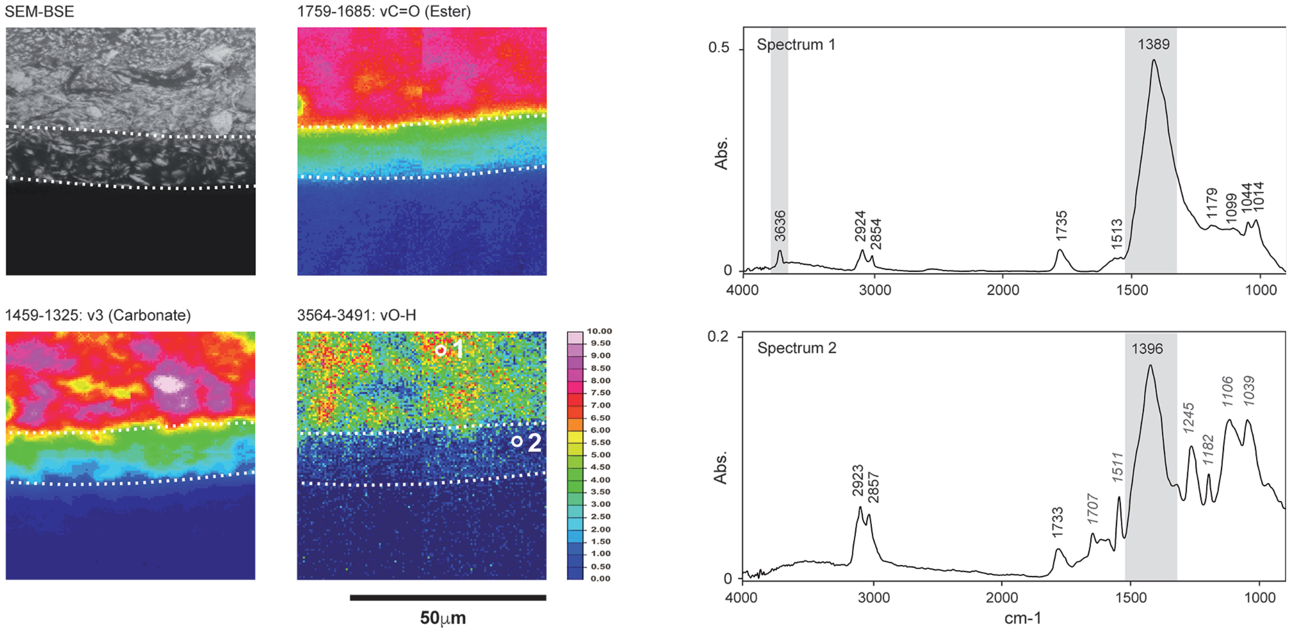

Figure 7. SEM-BSE and FTIR-FPA images of the outer layer at the delamination interface of sample S3. FTIR-FPA images represent the relative intensity of the corresponding spectral window denoted above each map. The numbers within the vO-H map correspond to the extracted spectra 1 and 2. Peaks with the grey background notify the signature of the lead white pigment. Peaks labelled with italic numbers signify the signal of the embedding resin. Illustrations: Zumbühl.

(Van Keulen 2014). The total ion current (TIC) chromatograms of both samples are almost identical in composition, although the paint flake has a higher relative concentration of palmitic, oleic and stearic acids. The TIC of the exudate in Figure 8 shows the distribution of fatty acid and diacid methyl esters. Peak numbers in Figure 8 are listed in Table 1. Mass spectra were identified by comparison to the mass spectral data atlas of J. van den Berg (2002), data from NIST (www.nist.gov) and by mass spectral interpretation. Note that oxy- and hydroxyl-groups are also methylated under the analytical conditions. In summary, a series of shorter- (C6-C10: nr 4, 5, 7, 9) and longer-chain fatty acids (C16-C24: nr 19, 22, 30), a dominant series of diacids (C4-C11: nr 6, 8, 12, 14, 15, 17, 18) with a Gaussian distribution, and several hydroxyl-, keto- and epoxy- stearic acids (nr 23, 24, 25, 26, 27, 28 and 29) were identified. The identification of glycerol (as methoxy derivatives nr 1, 2 and 3) points to preservation of some acylglycerol esters (as is corroborated by the FTIR data: Fig 6 D3). A preservation of the biological ester bonds would imply that the acid groups are mainly from the various diacids (formed by double bond oxidation) and much less by hydrolysis of ester bonds. Penetration in the alkaline lower paint layers, however, would certainly lead to hydrolysis of the ester bonds, thus making lead soap formation easier.

## Discussion

*Peinture 12 mars 1960* is a painting with severe damage due to weeping black surface paint and numerous areas of delamination and weeping black surface paint. The exudate shows characteristics of relatively high amounts of diacids and various oxy- and hydroxy-stearic acids in a free or glycerol ester form. We infer that these oil-medium-derived compounds not only appeared at the surface, but have also penetrated the underlying paint layers, where they reacted with the lead white mineral phases to form lead soaps. The diacid distribution from the surface paint shows a Gaussian distribution around the C9-diacid (azelaic acid), which suggests a thermal isomerisation of double bonds (Mills and White 1999; van den Berg 2002). The latest black paint layer is described by Soulages as the “black magma” created by himself with a mixture of ivory black pigment and linseed oil boiled with litharge (lead oxide) with addition of Flemish Siccative Medium from Bourgeois Ainé (Hélou-de La Grandière 2019: 394). Heating and boiling of vegetable oil leads to major chemical changes in composition due to thermo-oxidative stress (van den Berg 2002; Picariello et al. 2009). No evidence for resins was found in the GC-MS of the exudate or in the surface paint from *Peinture 12 mars 1960*, which made the addition of Flemish Siccative Medium questionable. In hindsight, GC-MS data of the lower-lying paint and ground layers would have been useful but imaging ATR-FTIR of cross sections has shed light on aspects of the chemistry of the layers.

The double ground of *Peinture 12 mars 1960* shows unique features that have not been reported before. The first ground layer is composed of coarse-grained lead white with some rounded barite particles, presumably used as extender. The second ground shows a relatively large amount of talc particles in a similar lead white composition. As the canvas does not have a stamp from the supplier, the question arises as to whether these grounds are indeed from a commercially-primed canvas of Lefebvre-Foinet, used quite frequently by Soulages by the end of the 1950s (Hélou-de La Grandière 2019: 395). This question remains open, requiring further comparative studies.

The first paint layer Soulages applied on *Peinture 12 mars 1960* is an off-white paint that consists of a fine-grained, well-sorted lead white typical for chemically-produced lead white (Corbeil and Sirois 2007). Delamination in the painting occurred exclusively where this paint layer was applied to the ground. The cross section in Figure 3 shows a relatively thick layer of degraded off-white paint where the lifting has occurred. FTIR mapping shows an abundance of lead soaps and oil binder components in this layer relative to the ground layers. The zone on the left has a different orientation of lead white particles, suggesting that underneath the lifted area the paint may have become more fluid. The morphology at the bottom of the paint flake from the major delamination zone shown in Figure 4 also suggests a difference in consistency when the lifting took place. The cross section made through an area with cohesion fillets of this paint flake (shown in Figure 5) makes the process during loss of cohesion visible in its distribution of the mineral particles from layers 2 and 3. The distribution of talc particles from layer 2 are mixed into layer 3, which is interpreted as a possible “fluidization” of the interfacial zone before or during the delamination process. Recently, evidence for a change in rheology at delamination interfaces has been reported by Bronken et al. (2019) on a delamination interface in

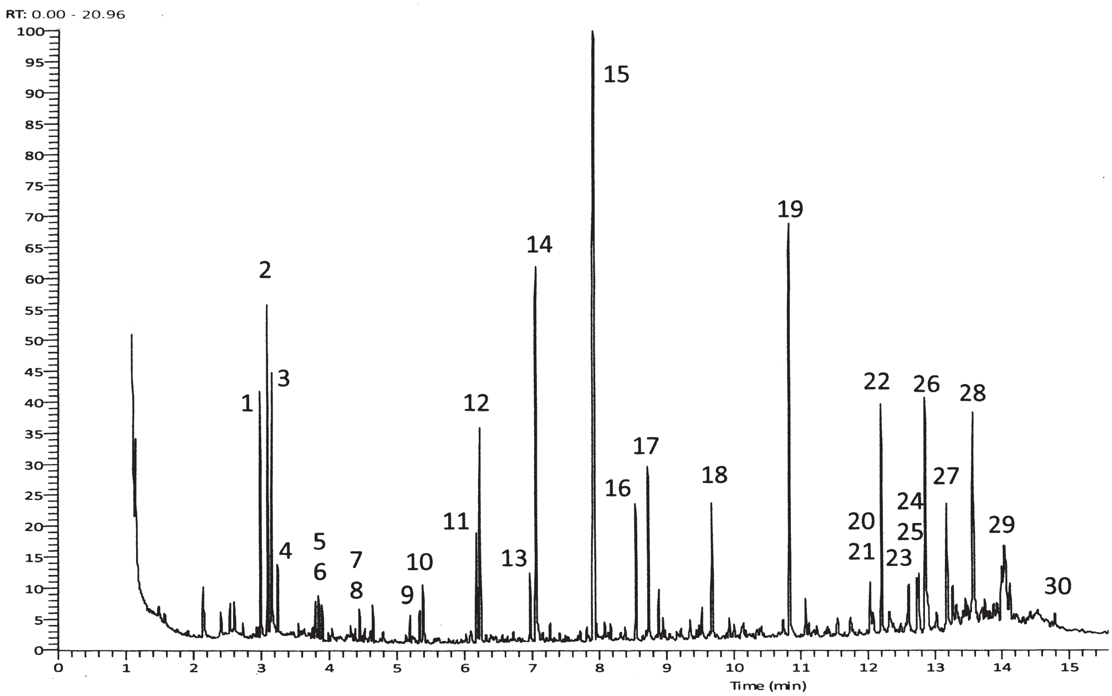

Figure 8. Total ion current profile of GC-MS data from the exudate. Peak numbers of compounds listed as identified compounds in Table 1 (right). Illustration: Boon.

a painting by Asger Jorn from 1957 and by Bronken et al. (2021) on a delamination plane in *Peinture, 10 Décember 1954*, by Pierre Soulages. More information on these interfaces in future studies should shed light on the dynamics of cohesion loss and subsequent delamination

The SEM-EDX maps and FTIR maps in Figure 6 illustrate a difference in chemistry between layer 3 and the new layer (2+3). There also is an ~10-micron-thin delamination zone present (Fig. 7) with a much lower density. Lead white (FTIR carbonate feature) and lead soaps are the dominant components in layer 3 and layer 2+3, but less so in the delamination interface of the cross section. The difference in binding medium components i.e., aliphatic features, acids and esters, and other lead soaps, can be observed, but the mechanism remains uncertain. The chemistry of the lead white in the delamination zone (Fig. 7) is different. Cerussite (lead carbonate) was found there while the layer above (2+3) consists of hydrocerussite (lead white) and plumbonacrite. This transition is indicative of less basic conditions in the outer zone during delamination. Our main conclusion is that loss of cohesion is not just a physical process of tearing but a much more chemically-reactive process of weakening at the delamination interface in the case of *Peinture 12 mars 1960.* It is inferred that a penetration of mobile medium constituents from the black surface paint plays an important role in the lead soap formation of the lower layers and the formation of a more fluid delamination interface. Physical forces subsequently lifted the paint at its weakest point. The cross sections in Figures 3, 4 and 5 illustrate the various stages of lifting.

| Number of identified compound in Fig 8 | Acronym (non-methylated) |
|---|---|
| 1 1,3-dimethoxy-2-propanol | GLY |
| 2 1,2,3-trimethoxy-propane | GLY |
| 3 2,3-dimethoxy-propanol | GLY |
| 4 hexanoic acid, methyl ester | C6 F |
| 5 heptanoic acid, methyl ester | C7 F |
| 6 butanedioic acid, dimethyl ester | C4 DF |
| 7 octanoic acid, methyl ester | C8 F |
| 8 pentanedioic acid, dimethyl ester | C5 DF |
| 9 nonanoic acid, methyl ester | C9 F |
| 10 hexanedioic acid, dimethyl ester | C6 DF |
| 11 8-methoxy-octanoic acid, methyl ester | omega OH/O-C8 F |
| 12 heptanedioic acid, dimethyl ester | C7 DF |
| 13 9-methoxy-nonanoic acid, methyl ester | omega OH/O-C9 F |
| 14 octanedioic acid, dimethyl ester | C8 DF |
| 15 nonanedioic acid, dimethyl ester | C9 DF |
| 16 tridecanoic acid, methyl ester | I.S. C13 F |
| 17 decanedioic acid, dimethyl ester | C10 DF |
| 18 undecanedioic acid, dimethyl ester | C11 DF |
| 19 hexadecanoic acid, methyl ester | C16 F |
| 20 octadecenoic acid, methyl ester (cis/trans) | C18:1 F |
| 21 octadecadienoic acid, methyl ester | C18:2 F |
| 22 octadecanoic acid, methyl ester | C18 F |
| 23 hydroxy-octadecenoic acid, methyl esters | OH-C18:1F (3 isomers) |
| 24 8-methoxy-9-octadecenoic acid, methyl ester | OH-C18:1 F |
| 25 11-methoxy-9-octadecenoic acid, methyl ester | OH-C18:1 F |
| 26 10-methoxy-8-octadecenoic acid, methyl ester | OH-C18:1 F |
| 9-methoxy-10-octadecenoic acid, methyl ester | OH-C18:1 F |
| 27 9,10-epoxy-octadecanoic acid, methyl ester | 9,10-EPOXY-C18 |
| 28 9,10-dimethoxy-octadecanoic acid, methyl ester | Di-OH-C18 F |
| 29 9,10-dihydroxy-octadecanoic acid, methyl ester | Di-OH-C18 F |
| 30 docosanoic acid, methyl ester | C22 F |

## Steps towards a conservation treatment

The painting has not been exhibited since 1986. The first serious defects in the form of lifting and dripping paint were noticed in 1995[6]. Since 1995, it was decided to record the condition very precisely, but not to treat the painting, in order to avoid a risk of successive inefficient treatments. This precautionary stance was useful in other similar cases and was justified since it guaranteed the ethical principle of reversibility. Unfortunately, during the two-decade-long period in storage in the museum in Tampere, the delaminated paint layer formed the open areas with curled paint seen in Figure 1A and Figure 9 in a process that appeared to accelerate in recent years. Treatment became urgent. After discussion with the curator, an experimental consolidation process could be started before all the information about the chemical process was available.

In order to stabilise and re-adhere the delamination, a compromise was sought to resolve the disparate behaviour of the paint film: the upper distorted paint was flexible while the paint remaining on the canvas was brittle. The aim was to re-adhere and bring the upper distorted areas back in line with the rest of the surface. The first step consisted of determination of the solvent and thermal sensitivity of both paint layers. Initial tests were carried out on the painted tacking margin. Only a few solvents were found to be safe to use without risk to the original paint layer: demineralized water; ligroin[7]; a solution of 70 parts ligroin to 30 parts alcohol (ethanol); and a 1:1 solution of water and alcohol. The paint was highly sensitive to raised temperatures (45°C is the softening temperature of the paint).

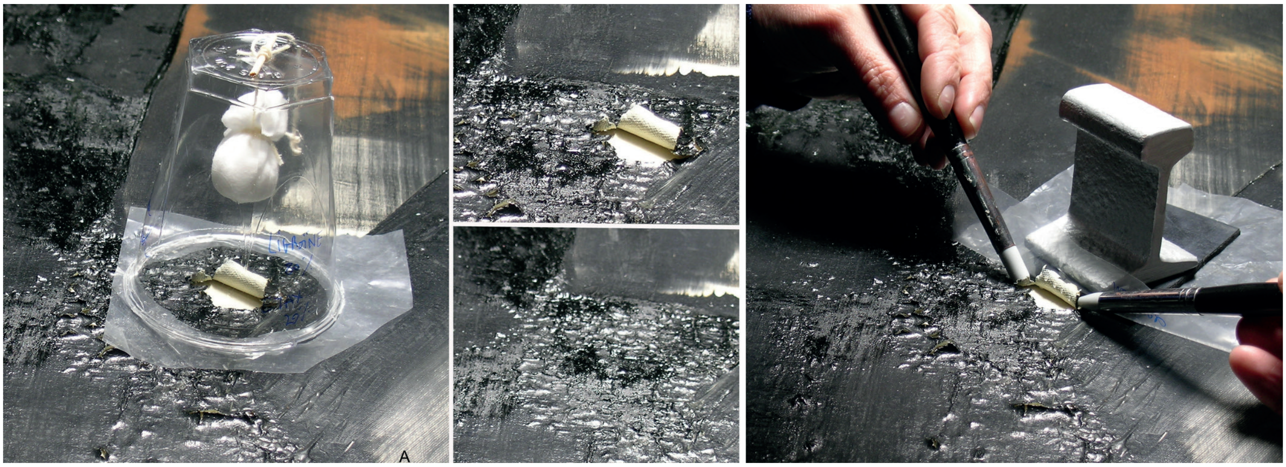

Figure 9. A short period of softening in an appropriate vapor atmosphere is sufficient to soften the curled paint for subsequent flattening (left- and right-hand side images). The image in the middle shows the before- and after-result of the repair. Photo: Hélou-de La Grandière.

The most challenging aspect was to flatten the somewhat malleable flakes. Figure 9 illustrates the chosen approach: in order to soften the paint while avoiding direct contact, the paint is exposed to solvent fumes locally by inverting a polyethylene cup over the targeted area, which contains a solvent infused cotton ball wrapped in fabric and suspended with string from the top of the cup. This micro-chamber delivers solvent vapours from a mixture of 80 parts ligroin to 20 parts ethanol. Only 10 seconds of exposure are necessary to affect softening in the paint, which retains flexibility for several minutes, allowing the curled flakes to be unrolled using a soft silicone spatula (a stiffer tool would mark the paint surface).

To select adhesives, tests done on the tacking margin revealed that few adhesives, which can be delivered in the limited solvents identified above, would adhere to the ground surface: Only Beva Gel® (Kremer), Klucel® G (in water or water/alcohol) and Lascaux Hydroground ® were suitable and effective (see Materials).

The consolidation was a compromise between good adhesive strength and an implementation compatible with the properties of the paint. An aqueous dispersion of Beva Gel® gave good results but was rejected because of its risky reversibility. Best results were obtained with Lascaux Hydroground ® (10% in water) plus a small portion of surfactant which strongly improves adhesion, probably due to a better wetting ability during the consolidation process (Triton® DF12, 2 ml in 200 ml solution). Previous experience supports this choice (Barabant and Hélou-de La Grandière 2012:326). The procedure involves two viscosities of the adhesive: diluted Hydroground ® in fluid form is inserted under the flake with a syringe and allowed to spread by capillary action. Then, a thickened formulation is applied under the flake (Hydroground ® thickened with 5 % by weight Klucel® in a 1:1 mixture with water and ethanol). Thanks to this thickened formulation, the adhesive does not spread beyond the lifted area, and its gradual drying can take place at the same speed as the flattening of the flake while it consolidates.

The paint is then laid down with heat (warm air from a Leister® gun) and pressure. The addition of Klucel® improves the viscosity and slows the drying time of the adhesive, while remaining compatible with the solvent. The initial tests using this consolidation method remained stable after 1,5 years, which is considered safe enough for further implement this treatment.

During flattening, the flakes appeared somewhat larger than the area of loss (Fig. 9) and required cutting the edges of original paint film by more than 1mm to reintegrate the flake into its original position: the white paint on the lower side of the flake apparently expands which explains the deformations of the paint impastos and curling. Due to this phenomenon, the flattened paint has lost its original thickness and surface texture; a process that is irreversible.  Although the “open wounds” could be healed, the paint surface is left somewhat disturbed and damaged. The modified consistency of the paint in its thickness is also an irreversible defect. The surface intended by Soulages can never be fully restored to its original characteristics, and the stabilisation of the painting can’t be guaranteed after consolidation.  This is an ongoing topic in the PhD project NOIRŒS.

## Conclusions

The investigation of the open surfaces of the delaminated areas on the painting provided physical and chemical clues to the process of loss of cohesion, cleavage and subsequent distortion in the remaining flakes. Ductile ridges on the delamination surface and supporting microscopic, SEM-EDX and imaging FTIR data point to different viscoelastic properties at the interface of Soulages’ off-white paint and the second ground layer below. The off-white paint and the second ground layer marked by talc crystals have lost their internal consistency and have mixed either before or during the delamination process. The composition of an exudate and its black source paint points to a thermally-treated binder that dried and matured but developed a non-cross-linked oxidised and acidic mobile fraction. This mobile material appeared at the surface but also penetrated the layers below. Part of this material has apparently been trapped as lead soaps, but another part is thought to have accumulated within the paint composite, causing a mechanical weakening at the interface of the artist’s off-white paint and the ground below.

Experiments to develop a novel treatment method were tested on the painting to flatten and re-adhere delaminated areas. While this procedure was successful in stabilising the paint, the surface appearance of the affected areas has changed, demonstrating that the effects of the paint degradation cannot be entirely reversed.

## Endnotes

1.The PhD NOIRŒS (acronym for Nouveaux Outils Interdisciplinaires pour la Restauration des Œuvres de Pierre Soulages / New interdisciplinary tools for the conservation of the works by Pierre Soulages) is a practice-led research doctorate in conservation affiliated with the Cy Cergy Paris University Graduate School of Humanities, Creativity, and Heritage. Under the direction of Thierry Sarmant, Mathieu Thoury (IPANEMA) and Lionel Simonot (Université de Poitiers), this work is supported by the Ecole Universitaire de recherche PSGS HCH Humanities, Creation, Heritage, Investissement d’Avenir ANR-17-EURE-0021.

2. « Pour rendre la toile plus glissante, il prépare une sorte de gelée ».

3. Personal translation of : « Soulages pose sur la table de marbre un bac rempli d'un magma noir d'ivoire. Il le trouve

trop dense. Il le rend plus fluide en ajoutant un liquide huileux. Dans un autre bac, il écrase un gros tube de blanc. »

4. « A la fin des années cinquante, j'ai eu, ainsi que quelques camarades qui avaient le même fournisseur que moi, pourtant très réputé, quelques toiles dont la couche picturale se désolidarisait du support »

5. EDX maps composed of several elements together representing one type of compound.

6. In 1995, painting conservator Kirsti Harva noticed exudation only from the paint layer. The drips were removed with a dry cotton swab.

7. aliphatic hydrocarbons without aromatics CAS Registry Number 8032-32-4.

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## Materials

- BEVA Gel: Aqueous dispersion of ethylene vinyl acetate and acrylic resins in a solution of water-soluble cellulosic material. Kremer 87032.
- KLUCEL G: Hydroxy-propyl-cellulose. Kremer 63706
- Lascaux Hydroground 750 : nBuMA-MA. Copolymer Isobutyl methacrylate, Methacrylate dispersion Lascaux 2001. Kremer 81027
- TritonDF12: non-ionic surfactant. Lascaux 4252

## Equipment:

Microscopy: Hirox KH 8700 with 2016Z lens (Hirox, Japan).

Fourier Transform InfraRed spectroscopy: FT-IR Focal plane array (FPA) imaging was performed on a Bruker Hyperion 3000/Tensor 27 infrared spectrometer with a germanium-attenuated total reflectance (ATR) crystal (20x objective A677-B10, crystal diameter = 250 µm). The spectral resolution achievable with the Cassegrain optics (NA 0.6) and the ATR Ge-crystal (Ge: nD 4.0) is between ~0.6 and 2.5 µm. The pixel resolution is 0.5 µm with the FPA detector made of 64x64 detector elements, covering an area of 32x32 µm (4 pixel per µm2). The 2D FTIR-FPA imaging was performed at 8 cm-1 spectral resolution and 64 scans (Spectral range: 4000-900cm-1).

Raman spectroscopy: Renishaw InVia (2007) system, 250 mm focal length, maximum resolution about 0.5-1.0 micrometer. Laser systems 785nm (Diode type), 633 nm (He-Ne-type and 514 nm (Ar-type). N-plan objectives on a Leica DM LM microscope.

Scanning electron microscopy: SEM-EDX: Zeiss EVO MA 10 SEM (2014) (W-cathode and variable pressure chamber) equipped with a Thermo NORAN System 7 for EDX measurements. Settings used for mapping were 20kV, 500pA, 512x384 pixel, 100 frames at 20 frames per second, (dwell time per spot 101µs) on carbon evaporation coated cross-sections.

GC-MS: The pyrolysis-TMAH-gas chromatography-mass spectrometry (Py-GC-MS) analyses were performed on a Thermo Quest GC-8000 equipped with a Supelco column Equity®-5, capillary GC column of 30m (I.D. 0.25mm, df 0.5) interfaced with a MS MD-800. See Van Keulen 2014.

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