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Transport and behaviour of water in oil paintings

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Chapter 7 Synthesis and outlook

This chapter synthesises the lessons learned in this thesis. Furthermore, it aims to place the findings into a wider context, by discussing the practical conservation implications and identifying three research lines for further study that may contribute to sustainable preventive conservation practice.

7.1 Findings in this thesis

This thesis initiated with an examination of chemical degradation phenomena in a case study from the 17th century in **Chapter 2**. Two paintings by Haarlem painter Jan de Bray were analysed with micro- and macro-imaging techniques. Both paintings contained a ground from which lead soap protrusions grew. Strong indications were found that liquid water exposure of one of the two paintings had accelerated or extended lead soap protrusion growth. Furthermore, detailed analyses of paint samples emphasised the complexity of historical paintings, in terms of their original materials, secondary formed products, conservation materials that have become an integral part of the painting, physical history and past environmental exposure. The case study illustrated that a holistic and interdisciplinary view is a requirement when studying cultural heritage materials.

Chapters 3 and 4 showed the potential of a simple water transport model based on ideal diffusion to generate insights into the behaviour of multi-layered paintings in dynamic environmental conditions. Validation against dynamic vapour sorption experiments and other experimental sorption data from literature confirmed that assuming ideal diffusion in ideal laminates is appropriate to describe bulk water transport in the layers of a painting on canvas. While there is considerable scope to expand the model to more accurately reflect the complex geometry and materiality of a painting, the main limit on the accuracy of computational results was found to be the parameter estimation. Data about the water sorption and diffusion properties of artist's materials, particularly aged materials, is scarce. Moreover, the parameter estimation based on gravimetric analysis performed in Chapter 4 showed that some variation is encountered in properties between seemingly similar materials. While this uncertainty limits the level of precision in the prediction of system behaviour that can be achieved with this modelling approach, the approach is still suitable for the investigation of the trends in behaviour of the layers of a painting, for instance in terms of response time. The model is a useful tool in disentangling the factors that determine a painting's response to the environment. We have shown that the response inside a paint layer, or any layer in a painting stratigraphy, depends on the sorption and diffusion properties and thickness of the layers, the stacking order of the layers, and importantly, the frequency and magnitude of relative humidity (RH) fluctuations. With so many factors acting together to determine the overall moisture response, the findings presented in Chapters 3 and 4 surpass our intuition of how paintings behave in fluctuating environments, hence showing the value of a computational approach.

The application of the water transport model to a case study of 18th-century painted wall hangings in the Hofkeshuis in **Chapter 3** showed that a gradient in RH caused by a temperature gradient between the front and the reverse of an oil painting on canvas can lead to a gradient in moisture content in the paint layers. A small average difference in RH (7%) between the front and the reverse was found to give rise a small absolute difference in water concentration between the first and second layer of paint. This gradient in moisture concentration correlated to a gradient in saponification of the lead-based paint layers in the wall hangings.

In **Chapter 4** the water transport model was employed to investigate the long-term performance of wax-resin lined paintings in relative humidity conditions following the BIZOT Green Protocol (40-60% RH, max. 10% RH change per 24 h). It was found that in the BIZOT conditions, the layers surrounding a paint layer in an unlined painting stratigraphy (varnish, ground layers, canvas support) shield the paint layer against humidity from the environment.

During the fastest allowed RH fluctuations under BIZOT conditions, the maximum water concentration in the paint layer was found to reach only 75% of the equilibrium water concentration at 60% RH. A wax-resin lining increased the shielding effect in these conditions, causing the maximum water concentration in the paint layer to reach only 30-40% of the equilibrium concentration at 60% RH. In the sensitivity study in Chapter 3, we learned that the moisture response is sensitive to the thickness of the layers. Therefore, the shielding capacity is not uniform throughout the painting, as the layers vary in thickness. Also, the frequency of the RH fluctuation is shown to be a major factor in determining the moisture response. Furthermore, we can conclude that wax-resin can undergo chemical change in 50 years of natural ageing in museum conditions, which results in increased water sorption properties. Further research is necessary to understand if ageing can also lead to increased water diffusion rates. By simulating different hypothetical scenarios of lined paintings, indications were found that changes in the properties of the wax-resin due to ageing may result in a more moderate response of the paint layer to RH fluctuations than expected. When the wax-resin impregnation does not seal the reverse of a painting fully and experiences an increased water diffusion rate due to ageing, water sorption into the painting occurs more gradual compared to a situation with an intact wax-resin layer, effectively buffering the response in the paint layer.

Chapter 5 showed that liquid water clusters can arise at the pigment-binder interface in water-saturated oil paint. This phenomenon was found to occur in oil paints with different mineral pigments, such as titanium dioxide, zinc oxide, lead oxide and iron oxide. The majority of water that is present in oil paint under water-saturated conditions, and all water at conditions below the saturation point, is present in the oil binder, either molecularly dispersed or in clusters small enough to be non-freezable. Only a small fraction of water in water-saturated conditions is aggregating near the pigment interface into nano-sized liquid domains that are freezable. This finding prompts us to consider aqueous chemistry inside oil paint.

Finally, **Chapter 6** tackled the challenge of monitoring chemical and physical changes in paintings using non-invasive techniques. Reactive model system that grew metal soap protrusion-like structures on a short timescale were employed for this study. For detecting changes in metal soap protrusions, 3D mapping techniques such as optical coherence tomography and microprofilometry were found to be suitable, provided that they reach sufficient spatial resolution in the plane of a paint layer ($\leq 20 \mu\text{m}$) and depth ($\leq 2\text{-}3 \mu\text{m}$). Furthermore, for comparing 3D maps in high detail, a robust spatial reference point is essential. More specificity for metal soaps is provided by techniques that couple high resolution 2D or 3D imaging to spectral information, such as microprofilometry coupled to VIS-NIR spectroscopy. Relatively low-tech techniques such as raking light imaging or commercial colorimetry do not provide sufficient resolution for detecting small scale changes associated with metal soap protrusions. X-radiography, routinely acquired in museums around the world, was found to be suitable for this purpose. Therefore, X-radiography with consistent acquisition parameters in combination with a form of automated recognition of protrusions could provide an accessible method for monitoring changes in protrusion distribution in affected paintings. 3D techniques with suitable resolution allow for more detailed change monitoring, not only in spatial distribution but also in shape and size of the metal soap protrusions.

7.2 Practical implications for conservation

The findings in this thesis suggest that chemical reactivity in oil paint increases non-linearly in high RH regions. This effect is caused in first instance by the non-linearity of moisture sorption isotherms of oil paint at high RH, where a strong increase in sorption occurs. In addition, when the RH is sufficiently high (likely >90% RH) or during liquid water exposure, local conditions near pigment-oil interfaces can arise with liquid-like water that can react, catalyse and transport species. While water-related chemical reactivity also occurs in moderate RH conditions, it will be accelerated when such local, high concentrations of water arise in the paint. This phenomenon may occur when an (unvarnished) painting is exposed to liquid water, for example during **active** (remedial) **conservation** treatment. Given the slow diffusion rate of water in oil paint, a typical exposure during conservation treatment would only result in water-saturated conditions near the top surface where water was applied or in craquelures. When considering **preventive conservation**, liquid-like domains inside the oil paint potentially arise in high RH conditions, when lasting sufficiently long. When a painting, wax-resin lined or unlined, is exposed to high RH for a few hours, the effect on the water concentration inside the paint layer is minimal, due to the shielding effect of the surrounding layers in the stratigraphy. Wax-resin linings continue to provide paintings with shielding against high RH conditions on the timescale of roughly a day. When high RH conditions persist on a timescale of days, the water concentration in the paint will rise too, which could eventually lead conditions in the paint that can accelerate the water-induced chemical reactivity.

7.3 Further research on impact of humidity on chemical reactivity

There are many potential next steps that can follow this research towards establishing a quantitative relation between chemical degradation of a painting and the environment. Another way to call this relationship is a dose-response function, which describes change in a material as a result of one or multiple factors, for instance relative humidity or temperature [1]. However, predicting chemical change in itself is not the endpoint. It should be coupled to macroscopic consequences to inform conservation decision-making. In addition, a layer of value can be added to a dose-response function that describes when change becomes unacceptable change, a so-called damage function. The attribution of value to change can be from an aesthetic point of view, or a more practical point of view, for example describing the loss of polymerisation in cellulose fibres in canvas supports to a point where the canvas cannot be handled safely anymore [2]. The development of damage function for oil paints is already an objective in the EU-funded project GoGreen (2022-2026) [3].

It is possible to extend the water transport model that was developed in this thesis with a dose-response relationship for chemical change in oil paint. A logical starting point for this research would be to establish a relationship between RH and hydrolysis in the oil paint polymer network, a reaction that essentially breaks down the binder in oil paint. As was discussed throughout this thesis, an oil painting can be considered a complex and heterogeneous system on many different length scales. Therefore, establishing a dose-response function for RH and hydrolysis is very challenging. A first step would be to classify the chemical response for oil paints with different types of pigments or driers. In addition, it would be interesting to further investigate the effect of liquid-like water clusters on reaction rates. As this type of research is commonly performed on physical model systems of paint, it is important to consider factors

such as oil pretreatment, pigment manufacturing and paint curing conditions to ensure relevance in relation to real works of art.

7.4 Condition monitoring as a parallel research line

The development of quantitative prediction of chemical degradation of oil paints, supported by damage functions and computational models, is ambitious. To reach a stage where they will inform preventive conservation decision-making is a goal for the longer term. Therefore, a vital parallel research line is the monitoring of real paintings. Condition monitoring is needed to understand if change is actively occurring and if so, at which rate. Condition monitoring will provide support to decision-makers regarding, for example, the adoption of new environmental policies, particularly when the consequences of such policies are not fully predictable yet. However, condition monitoring fulfils another function besides a flagging system that indicates active change. Condition monitoring is also a method to gain fundamental insights into change mechanisms and the factors that influence them. For this purpose, it is particularly insightful to monitor *unstable* objects, to gather information about when change becomes detrimental, or unacceptable. In a field where we would like to study everything, but have rather limited resources at our disposal, we must prioritise the study and prevention of this critical stage of change, i.e. the stage where damage actually occurs. Therefore, studying *unstable* paintings in *uncontrolled* environments are the key to more knowledge and insights about material change. This is the objective of the PREPARE project (2025-2029) funded by the Dutch Research Council [4]. Examples of uncontrolled environments are non-climatised historic buildings, in a variety of climate zones. Paintings in these environments are at high risk to climate change-related deterioration [5], and their institutions would benefit greatly from data to inform their sustainable decision-making. Furthermore, large-scale paintings found as part of historic interior decorations are interesting study objects, because they allow inter-painting comparison of different environmental conditions [6], an example of which is the collection of investigations centred around the painted wall hangings in the Hofkeshuis in Almelo, the Netherlands [7], [8], [9], [10], [11]. Altogether, to ensure impactful and relevant research into the critical behaviour of paintings, an intimate collaboration between conservators and researchers is key.

7.5 Redefining conservation

As part of research into conservation in the age of climate change and sustainability, it is necessary to engage in a wider discourse about change and lifetimes of cultural heritage materials, and importantly, our perception and expectations of these concepts. When does change become unacceptable to researchers, conservators, curators or members of the public? Do we have to develop a higher tolerance to change? While these questions are already an important part of conservation and conservation science, they will become increasingly inevitable in the future. Particularly in conservation of modern and contemporary art, reflecting on change has long been a central component in its practice that deals with inherently unstable materials [12]. In addition, as mentioned before, the unacceptability of change is a critical notion in damage functions and modelling lifetimes of collections [13]. Climate change is forcing the conservation community to address change more actively. In the built heritage context, frameworks about the management of change and loss are actively being developed and implemented [14]. However, we should be vigilant that concepts like loss management are

not solely applied to remote heritage sites that are difficult and costly to maintain, as this would lead to a further preferential loss of this type of cultural heritage. Sustainable conservation means performing interventions at a stage when they are most effective and allocating resources to prioritise the most vulnerable objects. Discussions about change and expectations are inextricable linked to sustainability and essential for the development and implementation of more effective and accessible approaches to conservation of cultural heritage in the future.

7.6 References

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