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Insights into the Effect of UV Radiation on Paintings: A Mini-Review for the Asset Preservation of Artworks

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HIGHLIGHTS

- A comprehensive insight into the photo-degradation process of paintings.
- Addressing a considerable role of the painting constituents in the degradation process.
- Appreciation of the preventive strategies to protect the heritage assets.

GRAPHICAL ABSTRACT



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ABSTRACT

The preservation of cultural heritage is of primary importance for human assets. The emphasis on painting stability has become an active area of research. The prolonged light exposure is a key factor governing the painting damage. This review addresses considerable insights into the chemical processes for the photodegradation mechanism. The paint formulation is the major enabler of chemical interactions with the photons in which the organic and inorganic materials react to generate free radicals.

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The formed radicals can proceed in chain reactions and cause an unacceptable change in the appearance or integrity of the painting. A thorough understanding of the degradation mechanism is dedicated to developing preservation strategies. Preventive measures are necessary to extend the longevity of the painting property. Restorers should be aware of the procedures of materials used in the painting practice, such as the compatibility of chemical composition, physical properties, and mixing ratios of the materials. The development of new materials could provide innovative strategies in the field of heritage conservation.

1. Introduction

Colors are incorporated into all aspects of life as a common perception and integral unit in human experience. The paint color is one of the main assets in archaeology and one of the main properties of materials [1,2]. The paint color arises from the absorption of specific wavelengths of light by electrons of the atomic structure of colorants. The human eye receives color in three attributes: hue, saturation, and chroma. The colorants may be natural or synthetic, soluble or insoluble, and organic or inorganic in composition to impart color to paintings. The binder is used to hold the colorant particles together and adhere them to the substrate material. There are many classes of binders, such as oil, proteinaceous, gouache, and encaustic. The paint film may be coated with a transparent protective and decorative varnish layer. Paintings represent a great proportion of the cultural heritage in an array of archaeological materials. Paintings are more vulnerable to color modifications over time as a change in the oxidation state of materials or the formation of new degradation products [3]. Color changes have a significant effect on the color balance of paintings.

The degradation of artworks is an intriguing phenomenon in the archaeological field. The decay of paintings is governed by many agents; ultraviolet radiation (UV) is one of the main contributors to the degradation process. The excited state properties of materials play a role in the chemical instability of painting. The strong UV radiation results in the formation of highly reactive free radicals or reactive oxygen species (ROS) that could cause decay of the painting layers, such as the superoxide radical anion (O_2^{\bullet}) , hydroxyl ('OH), alkoxy (RO'), organic peroxyl (ROO'), and hydroperoxyl radicals (HOO') [4]. The electron-transfer reactions can trigger a chain reaction mechanism and decay of the painting layers. In the field of heritage science, multi-analytical approaches can be applied to the examination of painting degradation. The main contribution of this article is to gain insights into the effect of UV radiation on paintings through a deeper understanding of the aspects of the degradation process and hence the role of preservation strategies.

2. Painting artworks

Paintings contain various components like colorants (pigments, dyes, or lakes), binders, and varnishes. The painting assets are often prone to damage and loss of value [5]. The degradation process is a complex subject that can be influenced by both internal materials used (via colorant/binder/varnish system) and external environmental agents (via light radiation) [3]. The degradation products could be responsible for the local molecular rearrangement of materials structure that induces the change of physical properties. In terms of light absorption, not all painting components have the same chemical response. Knowledge of the chemical structure of painting materials and their interactions with UV radiation is essential for understanding the degradation mechanism.

2.1. Degradation of painting materials

2.1.1. The entity of binder and varnish materials

When the binder (or varnish) materials absorb energy more than the bandgap energy, the organic radicals are produced (Fig. 1). The newly formed radicals react with oxygen to form organic peroxyl radicals that could react with another organic molecule to form hydroperoxide and alkoxy radicals [6]. The free radicals are capable of undergoing various secondary reactions that are independent of absorbed light. The radical species are



capable of abstracting a hydrogen atom and starting the auto-oxidation process. These species, in turn, produce oxygen-containing functional groups such as ethers, alcohols, esters, acids, aldehydes, ketones, and dicarboxylic acids. The photocatalytic reactions can promote the breakdown of the surrounding organic materials and can further cause the chemical alteration of surrounding colorants [7]. Sotiropoulou et al. mentioned that the auto-oxidation of fatty content of some binding media resulted in the formation of metal carboxylates as degradation products [8]. Yancheva et al. mentioned the infrared band at 1560 cm⁻¹ could refer to the presence of carboxylates that might be fatty acids-metal salts (fatty acid soaps) or resinmetal salts (resinates) [9]. The nanoindentation-atomic force microscopy (NI-AFM) was used to calculate the elastic modulus of samples taken from unaltered and altered painting areas. The higher values of the altered paint film contributed to the hydrolysis of drying oil and the increase in paint stiffness [10]. The presence of siccative pigments in oil-based paint could promote the surface drying of oil and accelerate the cross-linking and stiffness of paint film. Being a surface siccative, it induces significant changes in the mechanical properties via the formation of superficial wrinkles [11,12]. It has been suggested that the formation of α,β unsaturated ketones such as quinones is a major cause of the yellowing of light-aged varnish [13]. Scalarone et al. used gas chromatography-mass spectrometry (GC-MS) in conjugation with matrix-assisted laser desorption/ionization time-of-flight mass spectrometry (MALDI-ToF-MS) for the diagnosis of diterpenoid (colophony) and triterpenoid (dammar and mastic) varnish resins upon artificial light aging with a conclusion that the oxidation of the terpenoids and the incorporation of oxygen atoms per molecule [14].

2.1.2. The role of transition-metal pigments

Although metal-containing pigments are stable, they can catalyze photochemical reactions (Fig. 2) [15,16]. When the transition-metal pigments (via CdS, TiO₂, HgS, ZnO,

and Cr₂O₃ pigments) absorb energy equivalent to or more than the bandgap energy, the electrons jump to the excited energy state [11]. Specifically, the electrons (e) move into the conduction band (CB) and leave holes (h⁺) in the valence band (VB) [17]. The charge carriers (electron and hole) play a key role in the degradation process [18]. The conduction-band electrons react with oxygen adsorbed on the paint surface to form superoxide anion radicals, inducing the redox reactions. The valence-band holes react with a water molecule or hydroxide ion to form hydroxyl radicals, reacting with the organic binder [19]. The free water stored in the supra-molecular structure of binder materials (as a solvent in the preparation technique) has an effective role in which the water causes a rapid transformation of superoxide to hydroperoxyl and then the consecutive production of hydrogen peroxide and hydroxyl radical [20]. The stability of semiconductor pigments is related to the positions of the valence band maximum (VBM) and conduction band minimum (CBM) in relation to their thermodynamic oxidation and reduction potentials (φ_{oxid} and φ_{red}) and that of water [21]. The oxidation potential of the VB hole must be more positive than that of hydroxyl radicals (E^0 , $H_2O/OH^{\bullet} = 2.8 \text{ eV}$) to occur the oxidation reaction. The reduction potential of the CB electron must be more negative than that of superoxide radicals (E^0 , O_2/O_2 = -0.28 eV) to occur the reduction reaction [22]. The free radicals promote the chain-scission reactions, yielding low molecular weight species. The rate of chain scission correlates to the rate of radical production. The photodegradation processes continue to keep the decomposition process active. The termination step in the photodegradation process is the cross-linking in which different free radicals combine with each other [23]. The organic molecules decompose into small inorganic compounds (CO₂) and H₂O). Zhao et al. discussed the degradation process of a painting composed of tung oil binder and inorganic minium pigment (Pb_3O_4) under UV aging. The degradation process involved the reduction of the Pb(IV) into Pb(II) in red pigment and the oxidation of the ester/carboxyl groups in tung oil. The



<u>1- Chain initiation</u> B-H → **B**[•] <u>2- Chain propagation</u> B[•] + O₂ → **BOO**[•] BOO[•] + B-H → **B**[•] + BOOH BOOH → **BO**[•] + **•OH** 2BOOH → **BO**[•] + **•OH** 2BOOH → **BO**[•] + **BOO**[•] + H₂O BO[•] + B-H → **B**[•] + BOH B-H + BO[•] → **B**[•] + BOH <u>3- Chain termination</u> B[•] + BOO[•] → **BOOB** B[•] + B[•] → **B**-B B[•] + BO[•] → **B**-O-B

Fig. 1. Schematic representation of light-induced degradation process of binder in the painting system. Symbols represent the chemical species in the degradation pathway: B–H to the binder, B[•] to the binder radical, BOO[•] to the peroxyl radical, 'OH to the hydroxyl radical, and BO[•] to the alkoxy radical.

formed Pb(II) reacted with the obtained CO₂ and H⁺ to give a white product of hydrocerussite (2PbCO₃.Pb(OH)₂) with characteristic analytical features: XRD peaks at 24.8°, 27.4°, and 34.3° and Raman band at 1051 cm⁻¹ [24]. Van Driel et al. observed the chalking of the paint as a result of the photocatalytic activity of titanium white (TiO₂) pigment and the un-bounding of the binder at the paint surface [25].

2.1.3. The role of chromophores

Organic materials are fairly permanent and easily degraded in the painting system. Dyes containing covalently unsaturated groups are responsible for the color formation, also known as chromophores, such as -C=C-, -

C=O, -N=N-, -C=N, and -NO₂ groups. Other groups with non-bonding electrons can be attached to the chromophores to alter the ability of chromophores to absorb the light, and these groups are called auxochromes, such as -COOH, -COR, CH₃CO-, -CH₃, -NH₂, and –SO₃H groups [26]. When soluble dyes mix with an inorganic substrate (metallic salt), insoluble lakes are formed [27]. The chromophore groups present in the macromolecules can absorb the UV photons and initiate chemical modifications via radical intermediates formation. For instance, the varnishes are terpenoid compounds, and they are prone to chemical changes in the illumination conditions [28]. Zhuang et al. studied the interaction between alizarin (the main



 $TM + hv \rightarrow TM(\mathbf{e_{CB}}^{-} + \mathbf{h_{VB}}^{+})$ $TM(h_{VB}^{+}) + H_2O \rightarrow TM + \mathbf{OH}^{*}$ $TM(h_{VB}^{+}) + OH^{-} \rightarrow TM + \mathbf{OH}^{*}$ $TM(e_{CB}^{-}) + O_2 \rightarrow TM + \mathbf{O_2}^{-*}$ $O_2^{-*} + H^{+} \rightarrow \mathbf{HO_2}^{*}$ $B-H + OH^{*} \rightarrow \mathbf{New \ degradation \ products}$ $B-H + h_{VB}^{+} \rightarrow \mathbf{Oxidation \ products}$ $B-H + e_{CB}^{-} \rightarrow \mathbf{Reduction \ products}$

Fig. 2. Schematic representation of light-induced degradation process of transition-metal pigments in the painting system. Symbols represent the chemical species in the degradation pathway: TM to the transition-metal pigment, B–H to the binder, e_{CB}^{-} to the electron in the conduction band, h_{VB}^{+} to the hole in the valence band, 'OH to the hydroxyl radical, O_2^{-} ' to the superoxide anion radical, and HO₂' to the hydroperoxyl radical.

component of madder lake) and Al₂O_x film (aluminum oxides) under UV light exposure and noticed that the alizarin- Al_2O_x complex had led to an appearance of a new infrared band C=O group at 1705 cm^{-1} due to the oxidation of the catechol (C-OH) to carbonyl (C=O) group and the breaking of the conjugative connection between alizarin derivatives and Al³⁺ [29]. Machatova et al. employed the electron paramagnetic resonance (EPR) experiments for studying the UV photoexcitation of hydroxyanthraquinone dyes (HAQ) by using the spin trapping technique agent and the chemical quantum calculations. The photo-excitation process of hydroxyanthraquinones induced changes in the electronic absorption spectra that were linked with the ability of these compounds to generate reactive oxygen species upon light exposure [20].

2.2. Properties of paint formulation

Color permanence of paintings denotes that the pigments remain unchanged after prolonged exposure to light. The poor lightfastness could entail the darkening or fading of the paints. The painting color, in turn, depends on several material properties like particle size and refractive index of pigments [30]. Fine pigments (small particles) are associated with a higher binder demand during the dispersion process, homogeneous binder distribution, higher tinting strength, greater surface area, more light scattering, and hiding power. The coarse pigments (large particles) increase the surface roughness (inhomogeneous paint surface), the crack formation, and the surface area of the painting [31,32]. The larger surface area of the painting favors the chance of particulate matter deposition and accelerates the interaction with weathering agents [32]. In terms of the refractive index of colorants, the inorganic pigments have a higher refractive index than the organic pigments. The higher refractive index can scatter more light from the paint surface and decrease the light absorption by the pigments located in the painting. Paint gloss attributes to the pigment volume concentration (PVC), which is known as the volume of the solid particles (pigment and extender) to the total volume of solid com-



ponents and non-volatile part of the binder. The critical pigment volume concentration (CPVC) is defined as the point at which a sufficient amount of the binder enhances complete absorption with the pigment particles and close interstitial spaces in the paint film. The low wetting of pigment particles with the binder (higher CPVC) reduces the gloss effect of paint in a matt appearance, and this appears in the protrusion of pigment particles at the surface (tip of the iceberg effect). The high CPVC means there is less binder to fill the voids between pigment particles, and thereby direct contact of the painting porosity with the external environment via the humidity or pollutants. The CPVC value also has another effect on the painting degradation in which the higher amount of transition-metal pigments favors the more light absorption and the photocatalytic activity of pigments-mediated auto-oxidation [25]. The pigment load is decreased by adding fillers (or brighteners) to dilute and lighten the pigment. The addition of filler can change the surface appearance and modify the PVC value of the painting [11]. The binder demand in the painting technique is dependent on many factors, such as particle size, chemical composition, physical properties, and pigment-binder interactions [32]. As a consequence, the restorers should have knowledge about the working properties of painting materials.

2.3. Preservation strategies

Key attributes of light sources include the intensity of illumination, duration of exposure, and spectral distribution [33]. The photochemical action is equal to the product of the light intensity and the time exposure. Theoretically, there is no threshold of light intensity at which photo-activation will not occur. It is also worthwhile to mention that the lower illumination (light intensity or luminous flux) on the paintings can create fewer electron-hole pairs that participate in the decomposition reactions. It produces a fewer number of photons per minute but does not change their energy, which can play a role in photochemical deterioration. The shorter wavelength of the light source, the more energetic the incident photons over the painting surface and the more potentially damaging the object [34]. Preventive conservation is a mandatory proactive approach that consists of a set of policies and procedures to slow the deterioration rate of paintings via the control of the environmental conditions, maintenance. handling. packing. and transport procedures. Light control in museums, galleries, showcases, and storage areas are examples of preventive measures that provide a good contribution to painting stability. The preventive strategy is an ongoing monitoring process to avoid damage risks and to increase the life expectancy of heritage assets. Multiple scientific approaches could be applied to mitigate the chemical interactions with the objects. In the restoration practice, remedial or intervention conservation can be employed to improve the integrity of the painting and enhance the physical properties [35]. The synthesis of innovative materials can be a subject of future research to achieve the stabilization of the painting context [36]. The nanoparticles (NP_s) have recently been successfully applied in consolidation treatments [37]. Apparently, the engineering of the bandgap energy is a very important criterion for designing painting materials. The addition (or doping) of appropriate materials into pigments can enhance the electron-hole recombination and decrease the photocatalytic activity: broader bandgap energy, shorter life of holes in the valence band, faster recombination of electron-hole pairs, or more scattering light. Many surface coatings have been used to slow down the photochemical degradation process as a barrier to reduce light absorption and then the deterioration rate [38]. The coatings should fulfill the accepted standards of conservation via compatibility, transparency, reversibility, long-term lifetime, and nontoxicity [5].

3. Conclusion

The durability of paintings is markedly influenced by the surrounding environment. The uncontrolled environment has a harmful effect on the painting components. The intensive photons are a prerequisite for the electron transitions within the energy states of the painting materials causing physico-



chemical damage of paint layers. The protection of artworks is indeed a privileged task for the preservation of heritage assets where the preventive conservation/restoration treatments could reduce the impacts of photo-degradation at the earliest stages.

Conflict of Interest

The author declares that there is no conflict of interest.

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