Review

The use of erbium lasers for the conservation of cultural heritage. A review

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ABSTRACT

The characteristics of erbium lasers (Er:YAG) make them a promising tool for the conservation of cultural heritage, and yet they still remain less widespread than other lasers in this field. This review aims to summarise, compare and evaluate the results of case studies and experiments published so far about Er:YAG lasers for the cleaning of cultural heritage objects, such as paintings, stone, textiles, paper and plastics. The characteristics and cleaning mechanisms of Er:YAG lasers are presented. Research has focused on the application to painted surfaces and the damage threshold fluences of potentially sensitive pigments are summarised, along with those of organic substrates. The optimal irradiation conditions (fluence, wetting agent, pulse duration, frequency, etc.) for the removal of undesired surface layers, particularly varnishes, overpainting, encrustations and biological growth, are reviewed. This article also identifies the main achievements, limitations, potential applications and trends to foster research about the application of Er:YAG lasers in conservation.

1. Introduction and aim

The unique properties of lasers, such as intensity, monochromaticity, directionality and coherence, have favoured their use in the conservation of cultural heritage over the last decades [1,2]. Applications of lasers in conservation are diverse: chemical analysis (laser-induced fluorescence (LIF), laser-induced breakdown spectroscopy (LIBS), laser ablation mass spectrometry (LA-MS) or Raman spectroscopy), structural diagnostics (3D scanning, optical and digital holographic interferometry and Doppler vibrometry), optical imaging (Optical Coherence Tomography) and laser-removal of unwanted surface materials [1–3]. This review will focus on the laser cleaning of cultural heritage artefacts.

Laser cleaning was introduced in 1965 by Schawlow, who demonstrated the selective removal of black pigments from paper using a ruby pulsed laser [4]. In the early 1970s, the first attempts to remove encrustations from stone sculptures were made by Asmus on the portal of the *Palazzo Ducale* in Venice, achieving successful cleaning [5]. However, the limitations of the laser technology at the time (i.e. very low repetition rate, lack of appropriate systems to deliver the laser beam to the object, high cost and low performance compared with traditional methods), as well as the

cautious approach of the conservation community, practically confined laser technology to the laboratory for almost two decades. In the late 1980s further understanding of the process of laser cleaning (determined by both the laser parameters and the material properties), the development of the laser technology and the formation of an interdisciplinary community interested in lasers to solve cultural heritage problems, in great part thanks to the organisation of the LACONA (Lasers in the CONservation of Artworks) international conferences, led to the evolution of the field [2,3].

Today, although laser cleaning should not be considered a panacea for all conservation challenges, lasers are useful non-contact and environmentally friendly tools that offer great precision and control. For instance, laser cleaning has been extensively investigated and widely used for the removal of black encrustations from stone and also corrosion from metals, although there is discussion and ongoing research about the side effects, such as the laser-induced yellowing of white stones [6]. Neodymium lasers (Nd:YAG), emitting at 1064 nm and its harmonics, are the most commonly used for conservation purposes, being particularly successful for the cleaning of stone and plaster. Excimer lasers emitting in the ultraviolet range have also been applied to the removal of varnishes from paintings. However, erbium lasers (Er:YAG), used only since 2001, remain less established in conservation. Therefore, a review of the characteristics, success and limitations of Er:YAG laser cleaning is necessary to understand the potential applications and research gaps, and to foster research in conservation.

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2. Characteristics of the Er:YAG laser

2.1. Laser cleaning mechanisms

Er:YAG lasers emit radiation at 2940 nm and hydroxyl groups are the main absorbers at this wavelength. The selective absorption and subsequent heating leads to the explosive vaporisation of the OH-containing molecules in the treated surface, therefore **photo-thermal disaggregation** of the surface is the dominating mechanism. If the unwanted material does not contain OH groups, these can be added by applying a thin film of an OH-rich auxiliary liquid (i.e. water or alcohols) on the surface. In this case a **micro-distillation mechanism** takes place (with similar effects as steam cleaning). The OH groups of the liquid absorb the radiation and heat up to vaporisation. The rapid gas expansion leads to the disaggregation of the material on the surface (Fig. 1). The addition of an auxiliary liquid increases the absorption at the surface, favours a better distribution of the thermal load and confines the energy absorption to the top layer, creating a protective layer for the underlying material.

Er:YAG emits at a longer wavelength than most lasers (with the exception of CO_2 TEA lasers emitting at 10,600 nm). This implies a low photon energy (0.4 eV), which is not sufficient to dissociate most chemical bonds, so a photo-chemical ablation mechanism is not expected with Er:YAG lasers, as for example occurs with the UV excimer lasers (with photon energy of 4–6 eV). With this in mind, some researchers have investigated the chemical modifications in irradiated and ejected organic materials (varnishes and binding media) using gas-chromatography/mass spectroscopy (GC–MS) [7–17]. In general, laser irradiation induces no or minor chemical changes on organic materials, confirming that the mechanism involved in Er:YAG laser cleaning is physical. The only case of chemical removal mechanism reported in the literature concerns the removal of biological colonisation: de Cruz et al. [16] found that some compounds in lichens (polysaccharides and photosynthetic pigments from the algae component and lichenic secondary products) decrease in quantity or disappear following the laser irradiation. The laser radiation is therefore absorbed by OH-rich polysaccharides from the thick cellular walls of the lichen and the biological tissue consequently undergoes chemical breakdown [12,13,16]. The ablation of polysaccharides was the cause of white flashes observed upon irradiation.

The ideal scenario is the Er:YAG laser cleaning of OH-containing layers (organic coatings, biological growth, green copper corrosion, etc.) which absorb the laser radiation from hydroxyl free substrates which do not absorb Er:YAG radiation (i.e. stone, ceramic, metal, etc.). However, when vulnerable substrates are cleaned (i.e. paper, textiles, easel paintings, a secco wall paintings or with OH-containing pigments, etc.) additional care should be taken. The approach with the Er:YAG laser is usually to thin the unwanted layers rather than completely remove them. It is advisable to use decreasing fluences as the contaminant layer gets thinner and weaker and the sensitive substrate gets closer. In addition, recent developments in laser technology (lower minimum fluence and better beam quality) and in the knowledge of the damage threshold of the materials, may allow the exposure of paint layers with low risk of damage.

2.2. Material thickness removed per laser pulse

The -OH content of the irradiated material is a determining factor in its absorption coefficient for Er:YAG lasers. The high absorption coefficient of water and other OH-rich organic materials (such as many varnishes and adhesives), implies a shallow beam penetration, usually within a few microns.[1] Thus, theoretically, a thin layer of material is removed during Er:YAG laser cleaning, favouring a gradual and controlled process.

Optical tools, such as microprofilometry (μ-P), optical coherence tomography (OCT) and confocal Raman microscopy (CRM), were applied to monitor and evaluate the cleaning process in a non-invasive way and helped determining empirically the material thickness removed [11,20–23]. The experimental conditions and results are summarised in Table 1. In general, longer pulse duration values (i.e. 250–350 μs) and low fluences (<2 J/cm^2), with isopropanol as wetting agent, led to the removal of a thin layer (3–11 μm) of glue-based overpainting and acrylic resin [9,21]. When operating in the short free running mode (SFR) at about 100 μs and using higher fluences (c. 4–7 J/cm^2) the material thickness removed ranged between 10 and 30 μm for oxalate patina and shellac varnish, and increased to 100 μm with multiple pulses in synthetic resins [18,20,22]. Striova et al. [19] found laser cleaning more controllable and gradual to remove shellac from mock-up samples of murals than cleaning with solvents, as a laser pass left a residual 10–12 μm thick layer of shellac while a 10 min treatment with 5% ammonium carbonate left only a much thinner layer (<5 μm). Lorenzetti et al. [21] monitored the thickness of a layer of Paraloid B72 on mural painting samples before and after laser cleaning, which was useful to evaluate the cleaning efficiency of different experimental conditions [21]. They showed that the thickness of the layer, initially measuring between 6.6 and 10.4 μm, was reduced to approximately 3 μm after laser irradiation at fluences of 0.6 J/cm^2 in presence of isopropanol and disappeared completely at 1.3 J/cm^2. Operating at lower fluences (0.6 J/cm^2) with a mixture of isopropanol/acetone yielded a complete removal as well, due to the higher solubility of Paraloid B72 in acetone.

2.3. Thermal effects

De Cruz et al. [24] measured the surface temperature increase induced by the Er:YAG laser irradiation on a set of materials (samples from a panel painting, bronze, marble and limestone objects) with a purpose-built device, using graphite as a reference material and laser pulses with a fluence ranging between 0.1 and 2.6 J/cm^2. The temperature increase on the graphite was proportional to the average irradiation power. The tests showed that the temperature increase at the surface under dry conditions ranged between 45 and 140 °C, depending on the substrate and the irradiation power. Using a wetting agent stabilised the surface temperature and limited the temperature increase to only 15–18 °C, until the surface dried. This low increase is remarkable especially when compared with the high surface temperatures that can be reached with other lasers, such as the Nd:YAG laser, which can cause the melting of the substrate [3,25]. In relation to this, it is worth emphasising that the health and safety risks associated with the Er:YAG laser are lower than with the Nd:YAG laser, precisely because of the low surface heating and the shallow penetration in living tissue, as seen in the previous section.

2.4. Effect of the wetting agents

Several wetting agents are mentioned in the literature: distilled water, ethanol, mixtures of water and ethanol, isopropanol (pure or diluted in ligroin, water or acetone), water with polyethylene glycol (20) sorbitan monolaurate (Polysorbate 20 or Tween 20), diethylene glycol in white spirit and diluted ammonia solution. Research has shown that using wetting agents leads to a more efficient clean-

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[1] Thermal physics show that one single Er:YAG pulse at 1 J/cm^2 will vaporise c. 4 μm of pure water and heat to a depth of 5–10 μm depending upon the laser pulse duration.