

## E!2542 RENOVA LASER – Laser renovation of monuments and works of art

Jan MARCZAK, Marek STRZELEC\*, Andrzej KOSS, Rafał SZAMBELAN, Roman OKNIŃSKI, Matija STRLIČ, Jana KOLAR, Vasillis ZAFIROPULOS and Wolfgang KAUTEK

### Summary

*The paper contains a short description of the EUREKA E!2542 RENOVA LASER project and presents its aims, objectives, technological developments and market applications. It also describes selected examples of laser cleaning of various works of art, and presents the results of diagnostic tests of this technique's efficiency and accuracy.*

### Project description

The main aim of the project is the design, development and investigations of laser systems and technologies for the renovation of monuments and works of art (COOPER, 1998; ZAFIROPULOS, 1999; KOLAR 2000; KAUTEK, 2001; KOSS 2002). Diagnostic techniques for historic buildings and works of art, based on spectroscopic analyses of the laser-evaporated microparticles from the investigated samples, non-destructive surface testing using low-power lasers, and the visual inspection of an object's surface structure, are also under development. The particular objectives of the project have been as follows:

- Development of devices and techniques for the renovation of a wide range of historic relics, adapted to the material of the surface/substrate, as well as type, age and thickness of the surface encrustations.
- Development of laser and optoelectronic diagnostics of monuments, historic buildings and works of art.
- Development of integrated systems capable of controlling the amount of ablated materials.
- Development of a database for collecting the results detailing the efficiency and limitations of various laser-based techniques used in the restoration of works of art.

The solution to these objectives is based on several technological developments:

1. Optimisation and modification of the current laser sources:
  - Increasing the efficiency of the frequency conversion for the second, third and fourth harmonics of the fundamental Nd:YAG laser source wavelength and the development of the fifth harmonic generation (deep UV light).

- Improvement of the beam quality – a uniform energy distribution in transverse cross-section.
- 2. Adaptation of the acoustic process during the laser-material interaction, providing feedback to the surface exposure control during the laser cleaning.
- 3. Development of the surface diagnosis by illuminating it with a low-power laser and homodyne/heterodyne detection of backscattered radiation.
- 4. Adaptation of spectroscopy (laser-induced breakdown and fluorescence) for studying the chemical composition of removed surface encrustations (KLEIN, 1999).
- 5. Interdisciplinary discussion on the functionality of the laser equipment and its effectiveness, as well as the operating parameters for which laser treatment is user friendly and safe for the objects being treated.

Cultural heritage objects (buildings, monuments, statues, textiles, etc.) are cleaned conventionally by mechanical and/or chemical means, which provides limited control over the cleaning process. This can lead to the destruction of the surface layer and substrate, as well as create environmental hazards. Laser cleaning, in contrast, is a non-contact technique, which does not involve any chemical contamination or mechanical destruction of the objects. An expected effect of this project is the production and immediate introduction of laser equipment of competitive quality and price into service markets in Poland, Greece, Slovenia, Germany, Romania, and other countries.

### Examples of laser cleaning

Each individual work of art builds up its own characteristic surface layer, whose formation depends strongly on the substrate material and on the environmental conditions to which the artwork was exposed during its history. In many cases, cleaning is unavoidable, not only for aesthetic reasons but also to preserve the artwork. This is a complex problem requiring careful consideration of several parameters of the materials involved. The laser cleaning of artworks and historic relics has the advantage of a selective removal of undesired surface layers in a controlled manner and with a high level of precision. Sometimes, laser cleaning is the only way to remove encrustation from small, delicate works of art. The following results of the successful laser cleaning of

different and sometimes complex ancient works of art were obtained using a ReNOVALaser 2 device, based on the Nd:YAG, Q-switched laser with a maximum 0.5J of output energy, fundamental  $1.06\mu\text{m}$  wavelength, and minimum 6nsec of pulse width.

### ***Stone plates (MARCZAK, 2001/1)***

A very successful cleaning of high-relief stone plates was carried out. They were originally parts of balustrades in porches of tenement houses in Gdańsk, Poland. One of the plates was made in the second half of the eighteenth century and presents an allegorical figure with a panoramic view of Gdańsk in the background. It has the following dimensions: height – 72cm, width – 128.5cm, thickness – 12cm. Figure 1a shows the condition of the plate before cleaning. The value of the energy density was carefully selected during the laser cleaning process, in order to leave homogeneous, minute quantities of historic encrustation on the stone surface, forming a kind of patina that documents the authentic surface of the monument (Figure 1b).

### ***Limestone sculpture of Pantocrator (KOSS, 2001-1)***

The Tum Archcollegiate near Łęczycza is one of the most important preserved Romanesque architectural works in Poland. The particular fragment chosen is a limestone sculpture of Pantocrator, made in the eleventh/twelfth century and characterised by fine, millimetre-deep engraved drawings. A layer of gypsum and surface black encrustation was the result of the sculpture's long exposure to a polluted environment. The priceless bas-relief required a precise and non-invasive cleaning method which the selective laser ablation process could provide. The use of laser cleaning (Q-switched Nd:YAG laser) with the output energy below the predetermined threshold allowed us to remove all of the encrustation, while preserving the original substrate with a thin patina layer (Figure 2). A fibre optics spectrophotometer was applied to control the level of the limestone cleaning by means of measuring the backscattered reflection coefficient of white light radiation (see detailed description below).

### ***Terracotta (KOSS, 2001-2)***

Figure 3 presents a terracotta statue of St. Paul, the surface of which was covered by a thin, black and locally cracked layer of soil, formed due to prolonged exposure of the object to adverse atmospheric conditions. The laser beam was delivered to the object by means of an optical fibre with a collimating optical system. The output fluency was kept within the range of  $0.01$  to  $0.5 \text{ J/cm}^2$ , while the pulse repetition



**Figure 1.** Eighteenth century high-relief slab of a porch with representation of an allegorical figure with a panoramic view of Gdansk: a) before cleaning, b) after laser cleaning. Photo: T. Korzeniowski.



**Figure 2.** Pantocrator sculpture after laser cleaning.



**Figure 3** Terracotta statue of St. Paul with cleaned fragments.

frequency and pulse width were, respectively, 10Hz and 6nsec.

Investigations have shown that it was possible to attain extremely precise and controlled cleaning of the terracotta surface, which uncovered the pallid-pink substrate in its original state. Moreover, it was also shown that with the specific level of the fluency it was possible to:

- Clean the terracotta surface without damaging the protective, burnt layer.
- Clean the damaged terracotta surface without harming the underlying layers.

It should be emphasised that going beyond the determined laser fluency, even by a small value, increased the amount of the material removed. Moreover, the colour of the adjacent areas was observed to have changed from red to silver-grey. The latter effect was attributed to changes in the state of oxidation of the iron in the substrate surface as a result of its over-

heating. Long laser pulses (150 $\mu$ sec) were not used, since the high conductivity of heat to the depth of the material led to repeated vitrification of the terracotta surface.

#### *Ivory (KOSS, 2001-2)*

A particular difficulty in the cleaning of ivory is connected with its physical and chemical structure and the resulting properties. The hygroscopicity and anisotropy of ivory (inhomogeneous swelling in different directions) excludes the use of aqueous solutions. Moreover, acid and alkaline solutions are also not recommended. Experiments have shown that excellent results, without any damage to the ivory, can be achieved with laser cleaning utilising a wide range of laser fluencies.

#### *Alabaster (KOSS, 2001-2)*

Alabaster, a noble modification of gypsum, is a particularly fine-grained, white or slightly coloured mineral. Figure 4 shows a laser-cleaned alabaster flower-pot, made in the nineteenth century, which is the property of the Wilanów Palace Museum in Warsaw.



**Figure 4** Fragment of a laser-cleaned alabaster flower-pot.

**Flax gonfalon (HRYSZKO, 2001)**

The main problem in the conservation of old bunting is the frequent, simultaneous occurrence of metal threads and paint layers. It excludes the use of conventional cleaning techniques involving water solutions of thiourea and citric acid, followed by rinsing with water. Ultrasounds can destroy thin metal layers, and also require a water environment. In many cases, the only solution is to use low-invasive, dry laser cleaning.

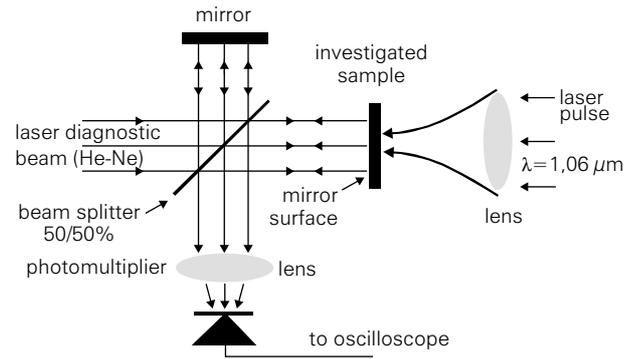
The successful laser cleaning of old fabric (flax) interleaved with silver and gold-coated silver threads, covered with paintings, was achieved using the ReNOVALaser 2 device with short pulses of 5-30nsec and 0.5J of output energy. The waste products of laser ablation were removed using small tampons and distilled water.

**Examples of diagnostic methods**

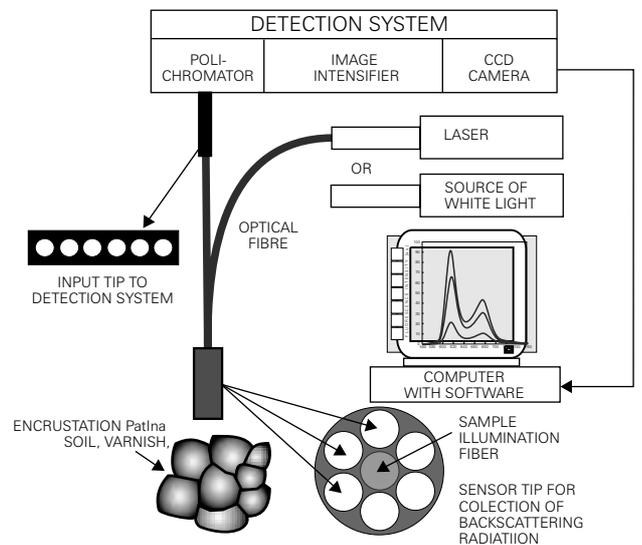
Laser cleaning is accompanied by several processes, also non-linear – such as dielectric breakdown, material evaporation and ablation, and thermally induced strains – generating elastic-acoustic waves. The generation and propagation of shock waves and subsequent acoustic waves along the surface or across the substrate can generate stresses, sometimes causing physical damage to the material surrounding the area illuminated by the high power laser beam. It can also cause damage to weak substrate material if soil is placed, for example, inside the surface cracks. Several diagnostic methods are being developed by the Polish partners of the project to monitor and control the efficiency and accuracy of this cleaning technique.

Pulse interferometry was found suitable for the measurement of the amplitude of the elastic wave generated by the pulse radiation of the Q-switched, Nd:YAG laser (STRZELEC, 2001). For this purpose, the back, mat side of one of the Michelson interferometer mirrors, made of marble, granite, etc. is irradiated with the focused, pulse laser beam. The front, polished side of this mirror serves as a totally reflecting surface for an He-Ne laser, which is a source of coherent light inside the interferometer (Figure 5). The variations in intensity of the interference fringes were detected using a photomultiplier and recorded by a TDS620 oscilloscope. The number of changes of interference fringes is a measure of the amplitude of the elastic wave generated by the laser pulse during cleaning. The increase of this amplitude is a warning against the possibility of damage to the object's substrate.

Figure 6 shows an experimental set-up with a fibre optics spectrometer for investigation into the reflection (scattering) coefficient of superficial layers (MARCZAK, 2001). As an example, Figure 7 shows the intensity of backscattered white light from ivory with a variable sur-



**Figure 5.** Experimental arrangement with the Michelson interferometer.

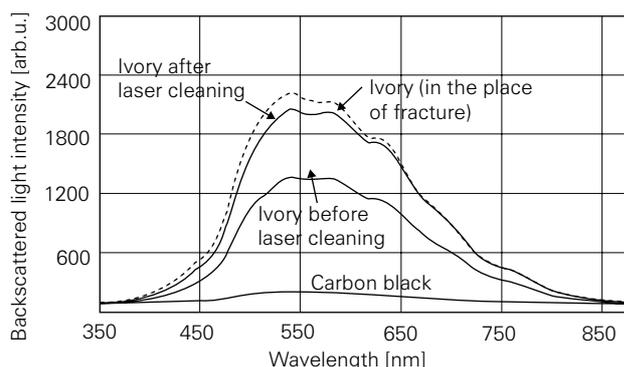


**Figure 6** Scheme of diagnostic system with a fibre optics spectrometer for investigation into reflection (scattering) coefficient of superficial layers.

face state (variable colour) as compared to the reference – carbon black. The use of a tuneable fibre optics spectrometer allows the selection of the most useful wavelength with the highest difference between signals obtained from the original and the cleaned surface. In the case of ivory, it is around 540nm. In the case of a stone plate, it is almost flat between 540 and 640nm. Utilising databases for reflection coefficients of different materials for various stages of cleaning, it is possible to estimate the progress of the process *in situ* and control it in real-time to stop the process before the laser beam interferes with the original substrate of the object.

**Influence of laser cleaning on the environment**

The two last meetings of the Management Committee of the COST Action G7 'Artwork Conservation by Lasers' were followed by workshops, which present-



**Figure 7.** White light backscattered from ivory and carbon black reference as the function of the wavelength.

ed several interesting papers concerned with aspects of safety and the effects of laser cleaning on air pollution. In our preliminary experiments, a simple glass cup enabled us to isolate the cleaned surface and to introduce a laser beam and a measuring probe. A sensitive opto-acoustic measurement system was able to detect small changes in gas concentrations at the level of ppm. Final concentrations of carbon monoxide and sulphur dioxide (well known as main pollutants of the atmosphere) can significantly increase, even double, during laser cleaning in closed areas.

## Conclusions

Experimental and theoretical work done during the first year of the RENOVA LASER project brought forth a lot of information fundamental to the understanding and optimising processes of laser renovation of different substrates with various encrustations. There is no space in this brief communication to present all of our conducted activities. Successive diagnostic systems based on laser-induced breakdown spectroscopy and laser-induced fluorescence are under construction. An Nd:YAG laser with the fifth harmonic ( $\lambda=212\text{nm}$ ) and an Erbium infrared laser will soon be operational. The results have been gathered in databases and will be used in subsequent experiments. The successful fulfilment of all our project objectives is expected, due to the high level of co-operation between participants.

## References

- COOPER M. (Ed), *Laser Cleaning in Conservation: an Introduction*, Butterworth Heinemann, Oxford, 1998.
- HRYSZKO H., KOSS A., MARCZAK J. and STRZELEC M., 'Laser cleaning of flax 17th century gonfalon', in *Lacona IV – International Conference on the use of Lasers in the Conservation of Artworks*, Paris, September 2001.
- KAUTEK W. and KONIG E., (Eds), *Lasers in the Conservation of Artworks II*, Restauratorenblätter – Special Issue, Verlag Mayer & Comp., Wien, 2001.

KLEIN S., STRATOUDAKI T., ZAFIROPULOS V., HILDENHAGEN J., DICKMANN K. and LEHMKUHL T., 'Laser-induced breakdown spectroscopy for on-line control of laser cleaning of sandstone and stained glass', *Applied Physics A*, **69**, 441–444, 1999.

KOLAR J., STRLIČ M., MULLER-HESS D., GRUBER A., TROSHKE K., PENTZIEN S. and KAUTEK W., 'Near-UV and Visible Pulsed Laser Interaction with Paper', *J. Cultural Heritage*, **1**, 221, 2000.

KOSS A., MARCZAK J. and STRZELEC M., 'Precise laser restoration of Pantocrator bas-relief at Tum's Archcollegiate – 12th century', in *Lacona IV – International Conference on the use of Lasers in the Conservation of Artworks*, Paris, September 2001/1.

KOSS A., MARCZAK J. and STRZELEC M., 'Review of preliminary work on laser renovation of different ancient objects in Poland – Part II (different substrates – small works of art.)', in *Lacona IV – International Conference on the use of Lasers in the Conservation of Artworks*, Paris, September 2001/2.

KOSS A. and MARCZAK J., 'Laser w konserwacji' (Laser in conservation), *Renowacje i Zabytki*, **2**, 76-81, 2002.

MARCZAK J., 'Surface cleaning of art work by UV, VIS and IR pulse laser radiation', *Proceedings of the International Society for Optical Engineering (SPIE)*, **4402**, 202–209, 2001.

STRZELEC M. and MARCZAK J., 'Interferometric measurements of acoustic waves generated during laser cleaning of works of art', *Proceedings of SPIE*, **4402**, 235-241, 2001.

ZAFIROPULOS V., GEORGIU S. and ANGLOS D., 'Lasers in Art Restoration', *Optics & Photonics News (OPN)*, 4-5 November 1999.

## Marek Strzelec\*

Institute of Optoelectronics,  
Military University of Technology  
ul. Kaliskiego 2,  
00-908 Warszawa, Poland  
e-mail: mstrzele@wat.edu.pl

Graduated in 1974 from Warsaw Technical University, received his Ph.D. in 1988 from the Military University of Technology (MUT). In 1976 he joined the Institute of Quantum Electronics of MUT, now the Institute of Optoelectronics. Since 1988 he has been an assistant professor. In 1993 he became the MUT representative at EXPO '93 in South Korea, as well as a consultant in the Korean Atomic Energy Research Institute. Coordinator or partner in several international projects. He is especially concerned with the physics and technology of CO<sub>2</sub> lasers and detection of infrared radiation. Member of SPIE, COST G7 Management Committee, the NATO Team of Experts for Chemical-Biological Defense and the Polish Optoelectronics Committee.

## Jan Marczak

Institute of Optoelectronics, Military University of Technology

### **Andrzej Koss, Rafał Szambelan**

Interacademy Institute for Conservation  
and Restoration of Works of Art,  
Academy of Fine Arts  
Wybrzeże Kościuszkowskie 37  
00-379 Warszawa, Poland  
e-mail: kossa@mercury.ci.uw.edu.pl

### **Roman Okniński**

COLT SA  
ul. Ostrobramska 75  
04-175 Warszawa, Poland  
e-mail: poczta@kolt.com.pl

### **Matija Strlič**

Faculty of Chemistry and Chemical Technology,  
University of Ljubljana  
5 Askerceva St., SI 1000 Ljubljana, Slovenia  
e-mail: matija.strlic@Uni-Lj.si

### **Jana Kolar**

National and University Library,  
Preservation Department  
1 Turjaska St., SI-1000 Ljubljana, Slovenia  
e-mail: jana.kolar@nuk.uni-lj.si

### **Vasillis Zafirooulos**

Laser & Applications Division, Institute of Electronic  
Structure & Laser (I.E.S.L.), Foundation for Research  
and Technology – Hellas (F.O.R.T.H.)  
Vasilika Vouton, P.O. Box 1527  
Heraklion 71110, Crete, Greece  
e-mail: zafir@iesl.forth.gr

### **Wolfgang Kautek**

Laboratory for Thin Film Technology,  
Federal Institute for Materials Research  
and Testing  
Unter den Eichen 87  
D-12205 Berlin, Germany  
e-mail: Wolfgang.Kautek@bam.de