applied optics

2D reproduction of the face on the Turin Shroud by infrared femtosecond pulse laser processing

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Femtosecond pulse laser processing concentrates a huge quantity of light energy in extremely short pulses of a few tens to hundreds of femtoseconds, enabling superficial laser machining or marking of any kind of materials, with a reduced or insignificant heat affected area. A digitized paper printed image of the face on the Turin Shroud was used to monitor a scan head intercalated between a femtosecond pulsed laser source and a linen fabric sample, enabling the direct 2D reproduction of the image of the face with a laser beam size corresponding to one pixel of the digitized image. The contrast in the marked image was controlled by adjusting the energy density, the number of superimposed pulses per pixel, and the distance between successive impacts. The visual aspect of the laser-induced image is very similar, at naked eye, to the source image. The negative photograph of the marked linen fabric reveals a face remarkably close to the well-known negative picture of the face on the Turin Shroud. Analyses by infrared spectroscopy, Raman spectroscopy, and scanning electron microscopy were performed to characterize the laser marked areas.

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1. INTRODUCTION

According to a paper by Di Lazzaro *et al.* [1], the inability to replicate the image of the Shroud of Turin, with all its microscopic details, makes it impossible to formulate a reliable hypothesis on how the body image was made on the linen tissue on which it appears. Some studies have obtained similar images, but without matching all macroscopic, microscopic, and chemical characteristics. Fanti [2] explored the hypothesis of image formation based on corona discharge. Such an energy source with its collateral temperature and UV radiation effects is considered by the author as one probable way the body image could have been formed. However, Fanti also said that no conclusion can be drawn concerning the mechanism that produced the image, because it is not possible to experimentally reproduce the source of energy needed to obtain a copy of the image on the original Shroud. Garlaschelli [3] asserted the reproduction of an acceptable replica of the Shroud using a rubbing technique applied to a human body, while the image of the face was obtained from a bas-relief to avoid inescapable wraparound distortion. However, Fanti and Heimburger (in a Letter to the Editor, [4]) contested some conclusions made by Garlaschelli wherein the replica obtained by the rubbing technique possesses all the features and the same visual and spectroscopic properties of the original Shroud. Ferrero *et al.* [5] reported that a CO_2 laser beam was able to reproduce the brown shades on linen fabric, with evidence of localized formation of tar on the surface of the linen fibers with a sponge-like structure due to swelling, but without any experimental evidence (as deduced from Fourier transformed infrared spectroscopy in attenuated total reflectance mode, FTIR-ATR) for the formation of carbonyl chromophore. The pattern of damage caused by the laser beam was similar to that obtained using an electron beam, even if in that case, the fibers were not linked in a sponge-like structure. However, heat treatment induced significant differences in the appearance and behavior of the fiber. Di Lazzaro *et al.* [6] investigated the use of ArF excimer laser radiation (193 nm) to obtain a color similar to that of the Shroud image by controlling the laser beam intensity and the number of pulses. These authors demonstrated that a short intense burst of directional UV radiation can produce colored linen exhibiting the many peculiar features of the Turin Shroud image.

However, to our best knowledge, femtosecond laser irradiation has never been used to reproduce the image of the Turin Shroud. Femtosecond laser generates light pulses lasting from a few tens to a few hundreds of femtoseconds. A review of knowledge and know-how on ultrafast laser synthesis and processing of materials was published in 2016 [7,8]. Figure 1 is a schematic diagram of the processing of laser–matter interaction, depending on the pulse duration, even if the differences in

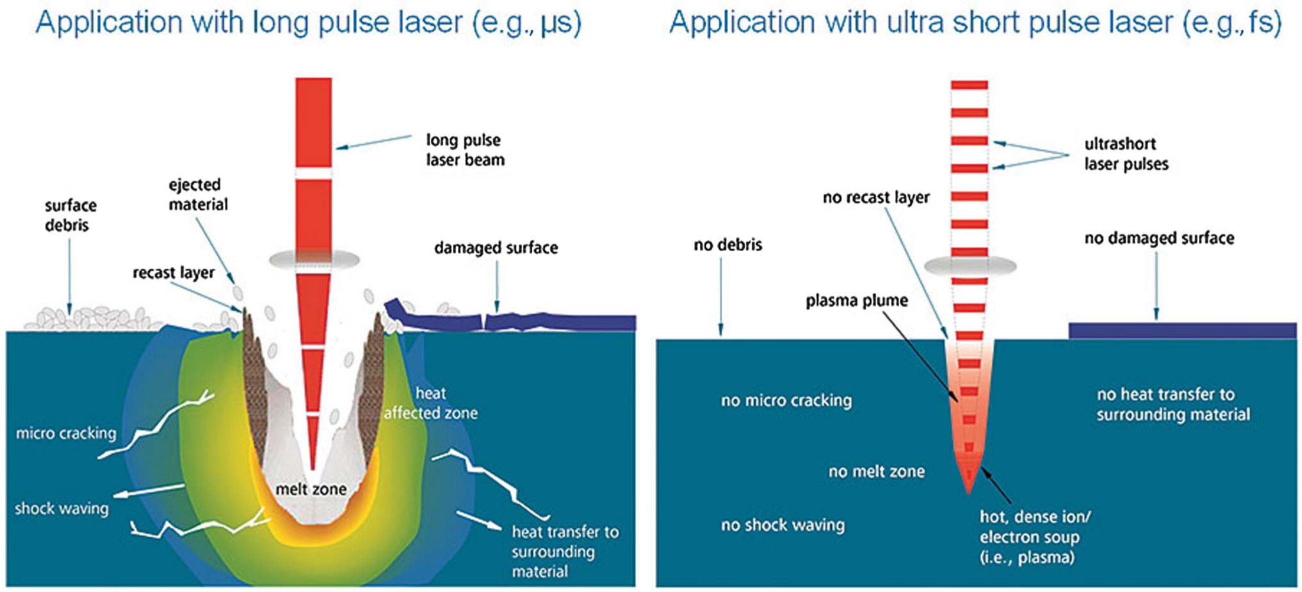

Fig. 1. Schematic representation of the laser–matter interaction, depending on the range of pulse duration. *Illustration courtesy of Amada Miyachi America Inc.*

the effects on the matter are not as marked as the scheme suggests (energy density in particular is also a crucial parameter in laser–matter interaction). In the sub-picosecond duration of the pulse, light generates free electrons that enable direct electron heating, non-thermal phase transformations, and a plasma state, depending on the nature of the material. Advantages over more conventional laser devices (delivering pulses longer than the typical few tens of picoseconds) are basically due to the extremely short time scale for energy absorption by free electrons of the irradiated material, with durations well below the electron–phonon coupling time (a few picoseconds) in most materials. As a consequence, thermal diffusion during laser interactions is said to be very limited, almost restricting laser energy dissipation to small optical penetration depths with minimal collateral damage. The use of femtosecond laser enables processing of practically any kind of material, including wood or fabric, with high precision and minimal collateral damage. Typical commercial femtosecond laser devices deliver infrared pulses with repetition rates in the kilohertz–megahertz range, with powers in the range of 10^8–10^{11} W per pulse.

The present proof shows, probably for the first time by using an infrared femtosecond laser, an image of the face on the Turin Shroud obtained by irradiation of a commercial linen fabric. The objective of the proof is limited to demonstrating that it is feasible, plus providing a precise description of the laser process. The results of preliminary basic investigations of the marked linen are also presented and discussed. An image of the face obtained by ultraviolet excimer laser was already obtained (see Fig. 4 in [9]). Thus, both images will be compared to comment the ability of two distinct laser processes, in particular in terms of wavelength and energy density, to provide a reproduction of the face on a linen fabric. The three main questions addressed in the present paper are as follows:

(1) What is the range of femtosecond laser parameters that can visibly mark a linen fabric without causing significant damage at the macroscopic scale?

(2) How can we combine the selected laser parameters with a scan head equipped with optical motors to process the laser beam from a digitized image used as the source of the marking?

(3) Is it possible to obtain an acceptable reproduction of the face of the Turin Shroud on a linen fabric by femtosecond laser processing consistent with a huge peak power per pulse (250 MW in the present experiment)?

2. EXPERIMENTAL

Commercial off-white cross-woven linen material, with a density of 0.24 $\mathrm{kg.m^{-2}}$ and a thickness of 0.3 mm, was used to perform laser marking. This density and thickness are close to the characteristics of the authentic linen Shroud of Turin (density between 0.20 and 0.23 $\mathrm{kg.m^{-2}}$ and thickness of 0.3 mm). The linen material did not undergo any preparatory treatment before the laser treatment. During laser marking, a flat piece of white paper was intercalated between the fabric and the sample holder.

A commercial paper printed image (size 22 × 15 cm) showing the negative of the Shroud face [Fig. 2(a)] was previously digitized with black-and-white contrast by a commercial scanner, to obtain a BMP image of 3000 × 2115 pixels [Fig. 2(b)]. Using the commercial GRAPHIC CONVERTOR software, this image was inverted to obtain a similar BMP image showing the positive Shroud face [Fig. 2(c)], as it appears on the original Shroud of Turin, except halftones not reproduced here due to the black-and-white digitalization (note that we consider the original Shroud image as a positive for our study, although it is in fact a negative with respect to the source from which it originates). The file related to Fig. 2(c) was used to form

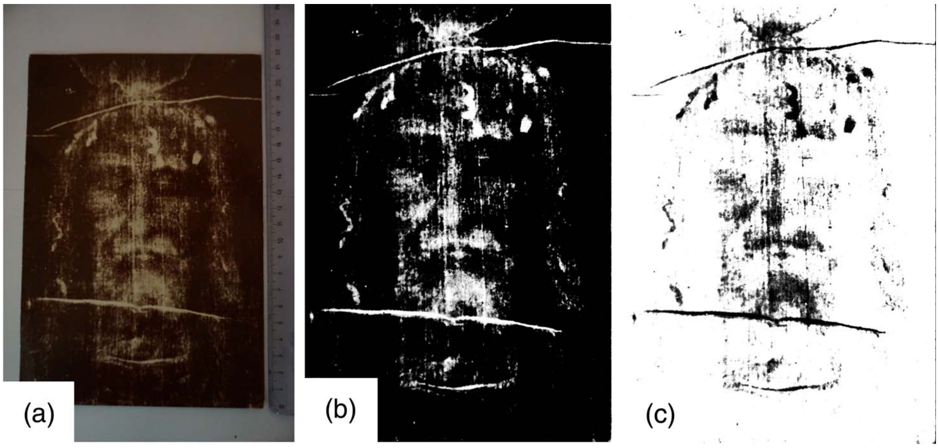

Fig. 2. (a) Commercial paper-printed image (22 × 15 cm) of the negative of the Turin Shroud face; (b) digitized image (3000 × 2115 pixels, black-and-white) of (a); and (c) negative digitized image of (b) used for the linen irradiation.

the image on the linen tissue using the laser process procedure depicted below.

The ytterbium femtosecond laser (Amplitude Systèmes) used for the experiment delivers 400 fs pulses, at a wavelength of 1030 nm, and a mean power of 2 W associated with a repetition rate of 20 kHz. This is consistent with an energy per pulse of 0.1 mJ and a peak power per pulse of 250×10^6 W. The number of delivered pulses can be monitored pulse by pulse.

As illustrated schematically in Fig. 3, the laser beam is introduced in a scan head containing galvanometer scanners made of high-precision rotary-motors for optical applications. To focus the beam, a lens with a focal length of 170 mm was fixed at the exit of the scan head. The laser beam, with a measured diameter of 40 μm focused on the plane of the sample, was scanned along the X and Y axes, with an adjustable number of pulses per pixel of the BMP image used as the source for marking.

FTIR absorption spectra have been recorded on a Thermo Nicolet iS10 spectrophotometer with ATR mode. Raman spectroscopy was performed using an Aramis Jobin Yvon spectrometer equipped with a laser emitting at 663 nm (1.87 eV) with a spectral resolution of 2 cm^{-1}. The laser beam was focused on the sample with a ×50 objective and the laser power was kept below 3 mW to avoid damaging the linen fabric. Scanning electron microscopy (SEM) was performed by a FEI field emission gun (FEG-SEM) at an acceleration voltage of 15 kV, into an atmosphere of 2 mbar of water vapor, in the “lens immersion mode” performed with a HELIX electron detector, to allow image formation with negligible charge effects.

3. RESULTS AND DISCUSSION

The contrast obtained from pulse laser marking depends in particular on the energy density, repetition rate, and distance between each juxtaposed irradiation impact. An energy of 0.1 mJ per pulse with a laser impact diameter of 40 μm corresponds to a mean energy density (or fluence) of 8 J.cm^{-2}. This value was kept constant throughout the experiment. The first experiment consisted in creating a contrast matrix (Fig. 4) with different laser impacted areas, whose visual appearance depends on both the number of pulses at a given $X-Y$ position corresponding to one pixel of the image to be digitized, and on the distance between each juxtaposed laser impact.

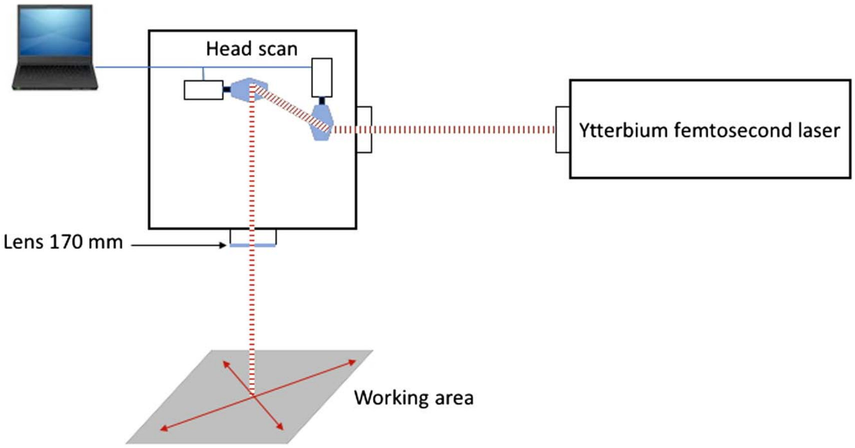

Fig. 3. Schematic view of the femtosecond laser device used for marking the linen fabric.

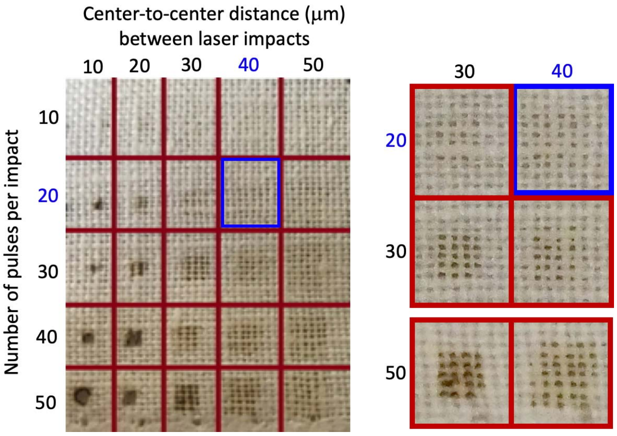

Fig. 4. Contrast matrix showing the impact of the femtosecond laser marking, depending on the number of laser pulses at a given $X - Y$ position, and the center-to center distance between juxtaposed laser impacts. The blue square corresponds to the conditions and appearance selected for the first laser marking of the face on the Shroud by femtosecond pulse laser.

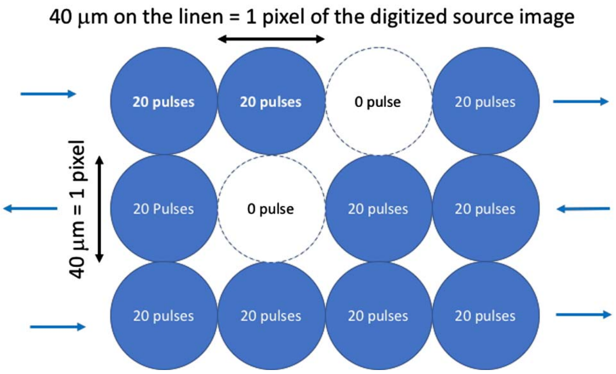

Fig. 5. Schematic representation of a representative part of the femtosecond laser impact matrix on the linen fabric. A dark pixel is marked with 20 superimposed laser pulses 40 µm in diameter. White pixels correspond to no laser impact.

The range of both process parameters depicted in Fig. 4 was selected from two extreme visual aspects observed on the linen fabric. A number of superimposed laser pulses as high as 50, with too much superposition between juxtaposed laser impacts (as low as 10 µm), leads to a too contrasted brown stain and severe damage to the fabric in the impacted areas. On the contrary, the same parameters, fixed at 10 laser pulses and 50 µm, respectively, result in invisible laser marking.

To obtain a femtosecond laser reproduction of the face on the Shroud on the linen fabric, we decided to select 20 pulses per impact corresponding to one "dark" pixel of the digitized image file, and 0 pulse corresponding to one "white" pixel of the digitized image file. The choice of a distance of 40 µm between the centers of two juxtaposed impacts each with a 40 µm diameter, in both the X and Y positions, was made considering (i) the visual appearance (Fig. 4, blue square), which resembles the moderate contrast of the original Shroud of Turin image, and (ii) the desirable order of magnitude of the size of the final reproduction (about 11 × 8 cm). Indeed, N juxtaposed impacts correspond to N pixels, forming a laser-induced dark line $N \times 40$ µm in length. The digitized image used to program the laser process was 3000 × 2115 pixels in the X and Y directions, respectively. As a consequence, the laser marked linen produced a 12.00 cm by 8.46 cm image. These dimensions correspond to the size of the marked linen sample, as shown below (Fig. 5). Finally, using this calibration, the laser processing was monitored to irradiate the linen tissue with a center-to-center distance of 40 µm between juxtaposed impacts corresponding to two juxtaposed pixels, 20 superimposed laser impacts for "black" pixels and no laser impact for "white" pixel, as schematically illustrated in Fig. 5.

Using our process, the image of the face on the Shroud was obtained in about 4000 s. A photograph of the image is shown in Fig. 6(b). A "1 euro" coin was placed on the fabric to give an idea of the scale. We observed a rather remarkable visual similarity between the face marked by the IR femtosecond laser [Fig. 6(b)] and the digitized image used to monitor the marking process [Fig. 2(c), duplicated as Fig. 6(a) to help easy comparison with Fig. 6(b)]. At the macroscopic scale, the laser marked fabric stood up to laser irradiation well, with no particular damage caused, no tearing of fibers. Based on the date the image was made (end of August 2018) and the time of writing (mid-October 2018), the image can be considered to be stable over this time period. By using the commercial GRAPHIC CONVERTER software, we obtained an inverted image of the marked linen, as shown in Fig. 6(c). This image corresponds to the negative of the photograph shown in Fig. 6(b), but is consistent with a positive image of the face of the Turin Shroud, as observed for the first time by the photograph Secondo Pia in 1898.

Details of the marked areas are depicted in Figs. 7(b) and 7(d), with squares identifying the diffuse but visible brown areas caused by the laser process. We observe that the inverted images depicted in Figs. 6(c) and 7(c) have a visual aspect very similar to the paper-printed image [Fig. 2(a)] used as the source for our marking experiment. Those observations do not imply that the IR femtosecond marked linen exhibits full similarities with the original face of the Shroud of Turin. Indeed, extended published results [10] and compilations [11,12] related to the Shroud of Turin image highlight a series of unique physical and chemical characteristics which have never been fully reproduced. Based on those considerations, one of the major basic characteristics, which needs to be investigated on a reproduction like the present one, is the chemical modifications induced on the marked linen by the IR femtosecond irradiation. Such preliminary investigations are based on FTIR and Raman spectroscopies, as detailed hereafter.

The results of ATR-FTIR analyses are depicted in Fig. 8, highlighting the spectral characteristics of virgin linen (black) and femtosecond laser marked linen (red). The spectrum of the virgin linen is consistent with data of the literature related to cellulose-based [13,14] and linen [15,16] compositions, with absorption bands characteristic of cellulose, i.e., the main constituent of the linen, and related to ν(OH) (broad band centered at 3335 and 1644 cm^{-1}), ν(CH) (near 2910 cm^{-1}),

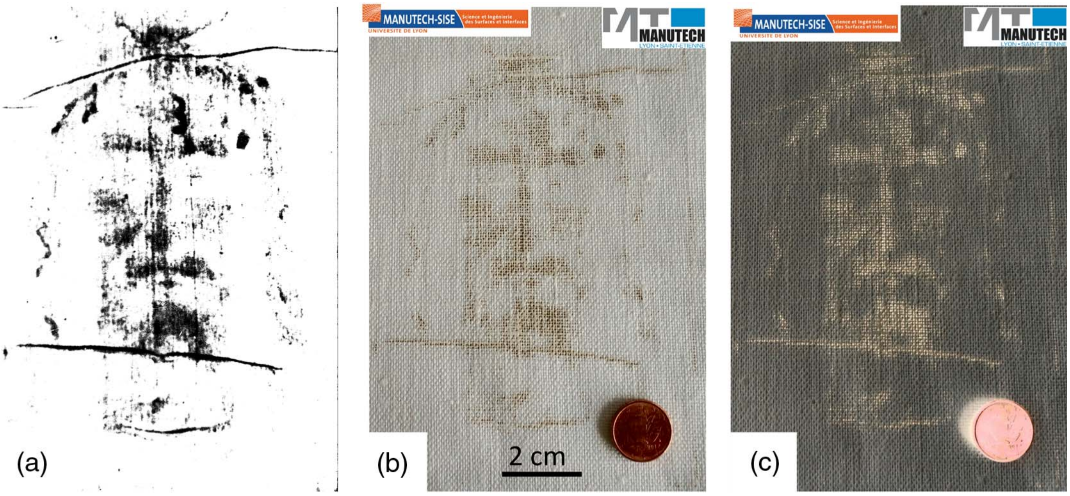

Fig. 6. (a) Same digitized image as in Fig. 2(c); (b) photograph of the linen fabric marked by the femtosecond laser monitored through the scan head from image (a); and (c) inverted photograph of the marked linen fabric depicted in (b).

$\delta(CH_2)$ (near 1432 cm^{-1}), $\delta(OH)$ (near 1315 cm^{-1}), $\nu(CC_{ring})$ (near 1112 cm^{-1}), and several symmetric bands corresponding to $\nu(C{-}O)$ in COH/COC groups (within 1200–900 cm^{-1}, and centered at 1037 cm^{-1}). The bands related to the femtosecond laser marked areas systematically resembled the spectrum obtained in the virgin area. In particular,

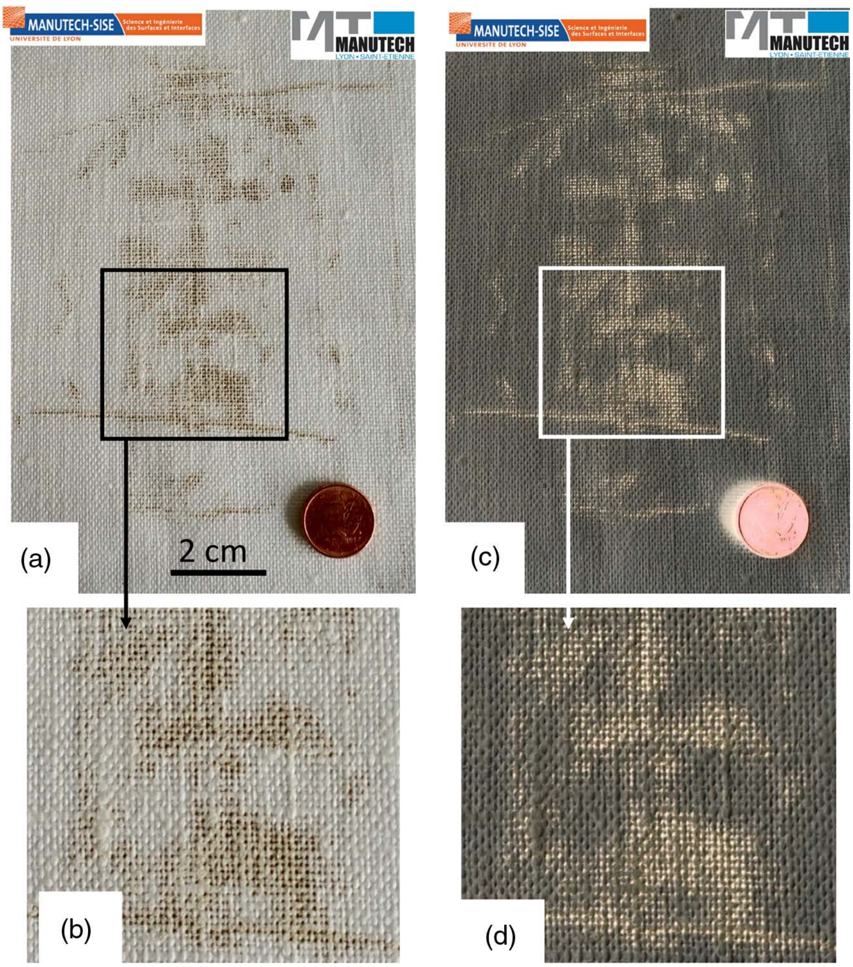

Fig. 7. (a) Photograph of the piece of linen fabric marked using the femtosecond laser with (b) details of the face; (c) negative of the image with (d) details of the face.

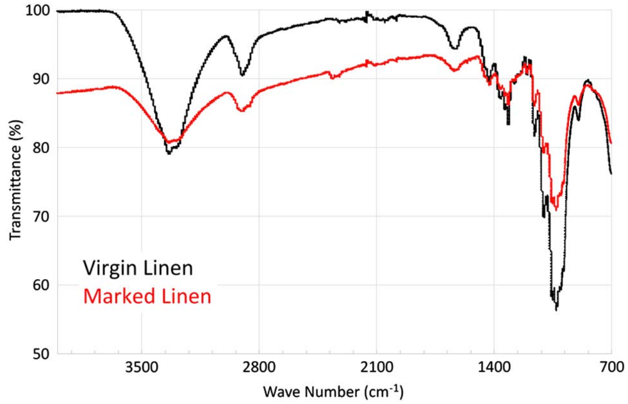

Fig. 8. ATR-FTIR spectra of virgin linen (black curve) and irradiated side marked linen (red curve).

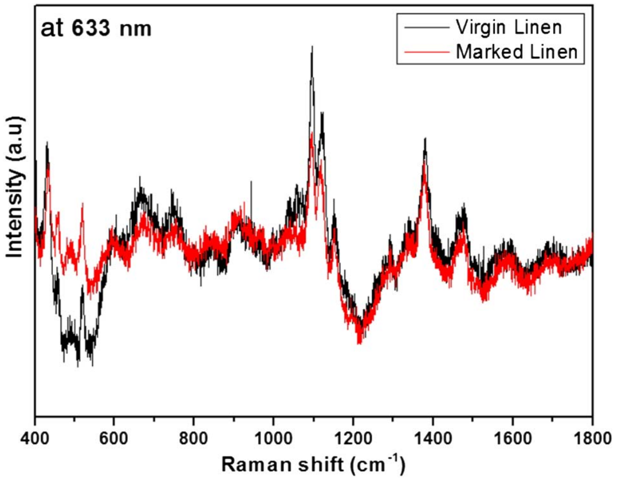

Fig. 9. Raman spectra of virgin linen (black curve) and marked linen (red curve).

no detectable C═O (in the 1680–1750 cm^{-1} range) that can cause yellowish or brown coloring due to laser-induced oxidation was observed on the spectra. The global transmittance of the femtosecond marked linen within 4000–1300cm^{-1} decreases compared to the virgin linen, in particular with significant decrease of the ν(OH) and δ(OH) absorption band intensities, consistent with some dehydration of the linen fibers which may be responsible for the weak coloration induced by the femtosecond laser irradiation. Indeed, according to Fanti *et al.*, as shown in similar ATR-FTIR spectra [17], the variation of the ν(C─O) in COH/COC groups (in the 1050 cm^{-1} range) and of the ν(OH) (in the 3350 and 1640 cm^{-1} ranges) are consistent with a dehydration process suffered by the marked linen fibers. These features are observed on typical image-fiber of the Shroud of Turin (see Fig. 23 in [17]). Moreover, if the C═O band (1720–1740 cm^{-1}) appears as a small shoulder of the 1640 cm^{-1} band in this figure, no similar shoulder appears in our FTIR analysis. Thus, the coloration of the image processed by IR femtosecond laser cannot be attributed to the formation of C═O chemical functions (at least with a sufficient contribution to be detected by ATR-FTIR), but more certainly to a dehydration mechanism of the fibers induced by femtosecond laser pulses.

Results of Raman analysis of the virgin and marked linen fabric are depicted in Fig. 9. The two spectra were superimposed after background subtraction. The main bands observed in linen are present, in agreement with [14]: δ(CCO) (near 435 and 455 cm^{-1}), δ(COC) (near 520 cm^{-1}), ring deformation (near 675 cm^{-1}), ν(C─O─C) (within 1100–1150 cm^{-1}), and $\delta(\mathrm{CH}_x)$ (near 1375 and 1480 cm^{-1}). As observed by FTIR, the two spectra are very similar. The marked linen Raman spectra does not exhibit significant new bands nor extinction of the bands observed with the virgin linen.

Kolar *et al.* [18] used FTIR to study cellulose-based papers irradiated by an XeCl excimer laser (17 ns, 308 nm, 16 Hz), by an Nd:YAG laser operating in two possible modes: (13 ns, 532 nm, 1.25 Hz) or (13 ns, 1064 nm, 5 Hz) at different fluences. These authors observed that cellulose irradiated with the 308 nm UV laser was subject to photo-oxidative degradation with a significant increase in the carbonyl and carboxyl groups. In contrast, at moderate fluences below 1 $\mathrm{J.cm}^{-2}$, the 532 nm laser had no effect. The 1032 nm laser-induced formation of inter- and intra-molecular cross-links, increasing the degree of polymerization, whatever the fluence ranging between 0.58 and 1.14 $\mathrm{J.cm}^{-2}$ at this IR wavelength. Thus, we cannot exclude the possibility that our IR femtosecond laser irradiation induces some polymerization of the molecules constituting the linen fiber, associated with some dehydration, but with no significant photo-oxidative reactions. Deeper investigations are required to elucidate the chemical origin of the brown contrast observed on our samples.

Figure 10 compares FEG-SEM images of the virgin tissue [Figs. 10(a)–10(c)] with images of representative areas of the marked linen [Figs. 10(d)–10(f)], at a magnification ×600, ×5000, and ×20,000. Virgin fibers are typical of linen whose yarn is made of agglomerated fibers with an individual diameter of around 20 μm. (Sub-)micronic heterogeneities are observed both at ×5000 and ×20,000. Such heterogeneities are not observed in [10], but the direct comparison of their optical micrographs with our SEM images is not obvious, except at ×600. After femtosecond laser irradiation, fibers are not cut nor broken, as observed in [4] after CO_2 laser treatment, but exhibit the appearance to have undertaken melting followed by rapid solidification, considering localized patches exhibiting a sponge-like structure, as highlighted at the highest magnification ×20,000. One cannot exclude, within these patches consistent with a re-solidified melted phase, a photo-induced polymerization process as observed in [18] with IR laser irradiation of cellulose. Deeper investigations using their approach are required to confirm this hypothesis with our experiment. Those investigations also require a quantification of the depth of coloration of the linen, which cannot be deduced from the present SEM analyses, nor by the FTIR and Raman results depicted above. Indeed, Fanti *et al.* [10] compiled the data related to the “superficiality” of the fiber coloration in the Shroud of Turin, and showed that the original linen contains a superficial chemically altered layer about 200 nm thick, the inner part remaining uncolored, consistent with a chemical alteration limited to the primary cell wall of the linen fibers. Probably, the

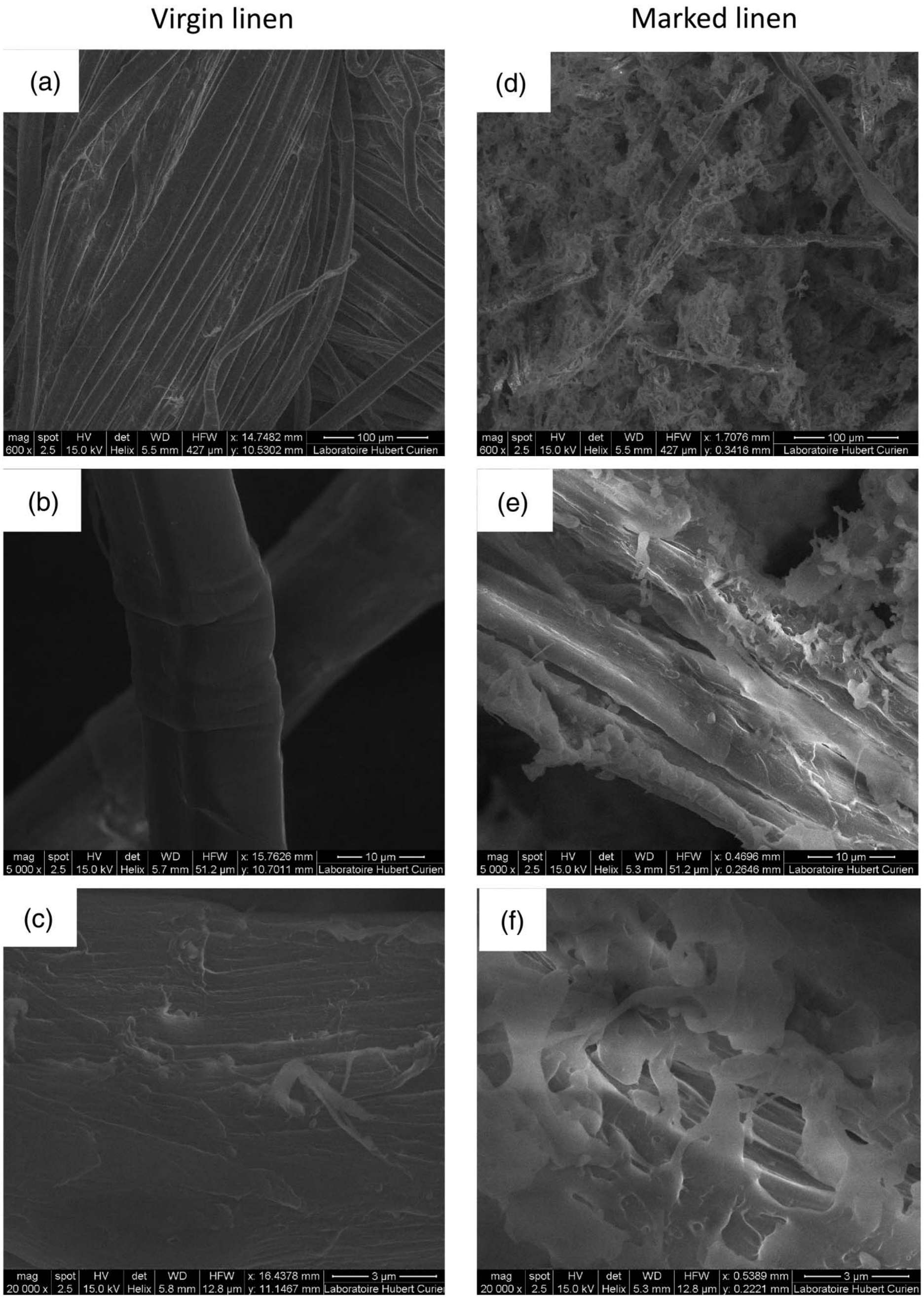

Fig. 10. FEG-SEM (immersion mode) of (a)–(c) virgin linen and (d)–(f) femtosecond laser marked linen.

infrared light of the femtosecond laser alters thicker than the first hundred nanometers, but this assumption requires deeper specific investigations.

4. CONCLUSIONS AND FUTURE DEVELOPMENTS

The present contribution provides some answers to the three main questions presented in the introduction. The first and main objective was to investigate the ability of an infrared femtosecond pulse laser source, coupled with a scan head, to reproduce an image of the face of the Turin Shroud on a linen fabric sample, and to analyze some chemical and morphological effects induced by the laser irradiation. The choice of the face of the Turin Shroud was motivated by the state of the arts of the scientific production related to this subject, exploring for the previous decades various energetic sources of radiation to explain how the original image was formed. We obtained, probably for the first time by using an infrared femtosecond laser, a 2D reproduction of the face on the Shroud of Turin. The laser was combined with a scan head delivering a controlled number of pulses (40 μm in diameter) juxtaposed at a controlled distance corresponding to their diameter. The source was a BMP file containing the image of the face on the Shroud, and each pixel of the image corresponds to a position of a laser shot. The visual appearance of the irradiated linen, as well as its negative photograph, offer an image with some resemblance, in terms of visual appearance, with the original picture of the face of the Turin Shroud. The image has a similar aspect as the face obtained by the ultraviolet excimer laser (Fig. 4 in [9]), but the spatial resolution obtained by the infrared femtosecond laser looks better. This demonstrates that an infrared femtosecond pulse source of light is able to produce an image on the linen fabric, mainly based on a dehydration mechanism of the linen fibers. Even if the femtosecond laser is known to induce limited thermal and chemical effects on irradiated materials, including organic ones, our preliminary investigations indicate that such alterations, at the microscopic and chemical scales, are significant enough to induce the formation of an image on a linen fabric sample.

Efforts can now be invested in the optimization of the process and deeper investigation of the analyses, in particular:

- The effect of variations in laser process parameters (fluence, beam size diameter, distance between laser shots, repetition rate) can be explored around the conditions we chose for this prototype experiment. The coloration intensity (in particular by considering halftones) and coloration depth inside the marked linen need to be systematically investigated, depending on the laser parameters.
- The links between the laser process parameters and the resolution of the image need to be systematically investigated, in particular to produce an image whose dimensions are the same as those on the original face on the Shroud of Turin.
- Even if the remarkable stability of the FTIR and Raman signatures after femtosecond laser irradiation is not consistent with a significant photo-oxidation process, but rather with some polymerization of the linen compound, this requires further investigation.

In agreement with the scientific community investigating the fascinating question of the formation of the image on the Shroud of Turin, the perfect reproduction of the Turin Shroud remains a challenge since many other characteristics of the original image have not yet been obtained with any of the processes used for reproduction to date.

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