Experimental study of the effect of a laser beam on the morphology of wood surfaces

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ABSTRACT

The study reports on fundamental aspects of modification of wood by using a laser beam and relates different parameters such as laser type, intensity, time and focus to the surface morphology that presents ablation or melting processes that can be seen from microscopic observations (SEM). The penetration of water into laser treated surfaces is considerably suppressed. Apart from ablation or sealing of the wood surface, the laser beam can be used to remove microscopical amounts in order to clean or to prepare the surface for coatings by controlling the energy input. The paper shows some applications in the preservation of historic monuments. It is thought that the modification of wooden surfaces by laser beam represents a fundamental technology with many potential applications.

Keywords: wood, cellulose, lignin, laser, ablation, melting, cleaning, infrared, ultraviolet, self-limiting process

1. INTRODUCTION

Wood is a porous material whose porosity amounts from 60\% to 70\% for coniferous wood. The physical characteristics of wood, especially its moisture behaviour, strongly depend on the anatomical aspects. It consists of different chemical compounds among which cellulose, hemicellulose, and lignin are the most important ones. These chemical compounds consist of polymer chains that do not show a precise melting point but a glass transition temperature. Its value is distinct for lignin and cellulose and varies between 140°C and 200°C respectively \cite{1,2}. Therefore, “melting” of wood without burning or blackening of the surface cannot be achieved with conventional heat sources.

![Figure 1. The laser technology permits a precise control of the energy input which can be used for machining of wood and allows modifications on logs (a), group of cells (b), cell wall layers (c) and the fibrillar structure (d).](image)

However, the laser technology permits a precise control of the energy input which can be varied by changing the wavelength of the beam (CO\textsubscript{2}, Nd:YAG, excimer), puls duration, repetition rate, and intensity so that the anatomy of the wooden surface can be modified within the range from laying bare the cellular structure to sealing the surface by melting and solidification.

Seltman \cite{3} was the first who published results of ablation experiments on wood surfaces by excimer lasers. The aim of those investigations was the ablation of the thin sheet of damaged wood structure due to mechanical processing in order...
to improve the adhesion of glue and protective coatings. The thickness of this layer (typically 60 µm) depends on the quality of the tool edge and on its pressure on the surface. The surface structure obtained by ablation was similar to that of a microtome cut for SEM investigation.

The study reports on fundamental aspects of these phenomena and relates different parameters such as wavelength $\lambda$, power density $E$, pulse duration $T_p$ and focus to the surface morphology that presents ablation or melting processes that can be seen from microscopic observations (SEM). The penetration of water into laser treated surfaces is considerably suppressed.

Apart from ablation or sealing of the wood surface, the laser beam can be used to remove microscopical amounts in order to clean or to prepare the surface for coatings by controlling the energy input. The paper shows some applications in the preservation of historic monuments. It is thought that the modification of wooden surfaces by laser beam represents a fundamental technology with many potential applications.

2. EXPERIMENTS ON LASER ABLATION [4,5]

Orech [6] measured the absorption spectrum of wood. Because of the absorption minimum at 1000 nm the lowest ablation per pulse is expected for Nd:YAG lasers. Contrary to this, an effective ablation can be predicted for the UV- and IR-spectral range considering an absorption of > 80 % in this spectral range. Therefore, the investigations were concentrated on experiments in UV and IR.

The small effect of 1064 nm laser light on wood was shown by simple ablation experiments with an Nd:YAG laser (NY82S-10 Continuum, 1064 nm, 8 ns, 1.8 J).

2.1 Experimental Setup

Regarding future technical applications, a XeCl-excimer laser (XP2020, Siemens, 308 nm, $T_p= 40$ ns) and a CO$_2$-TEA-laser (Uranit, Urenco, $\lambda = 10.6$ µm, $T_p= 1.2$ µs) were used for experiments. For ablation by excimer laser a beam guiding system with homogenisation and imaging of the smoothed intensity distribution by an objective lens was used. In those experiments, the power density on the wood surface was varied between 0 and 25 $10^6$ Wcm$^{-2}$ by help of an attenuator. For ablation in the IR-range the laser beam of the CO$_2$-TEA laser focused by a 45° focusing mirror of molybdenum coated copper with a focal length of 400 mm was used. To change the power density between 1.7 $10^6$ Wcm$^{-2}$ and 4.2 $10^6$ Wcm$^{-2}$, the distance between sample and mirror was varied.

Wood surfaces oriented according to the principal cut orientations (cross - across the trunk, quarter - radial, slash - tangential to the growth rings) from pine and beech have been investigated.

The excimer laser ablation process was in-situ observed by a high-speed camera system (IRO-Image Intensifier Camera, PCO-Computer Optics). The reached ablation was measured by a perthometer (S3P/PRK, Feinprüf Perthen GmbH).
2.2 Results for ablation experiments

As found for artificial polymers like polyimide, an power density of 25·10^6 W cm^-2 at 308 nm is already sufficient for ablation of wood of any species, orientation and moisture. Figure 4 a,b,c shows SEM images of ablated beech wood surfaces. The inner structure of the wood is exposed to the view after removal of the squeezed surface layer by laser ablation. Similar results were found for pine, although the microstructure was different. Ablation occurs in the same way with IR irradiation. For suitable parameter sets, damage-free ablation of the wood structure without carbonisation is possible.

Figure 4. SEM-images of laser irradiated surfaces of beech wood in cross (a), quarter (b) and slash cut (c). Irradiation parameters: \( \lambda = 308 \text{ nm}, Q = 0.9 \text{ J}, T_p = 40 \text{ ns}, f = 5 \text{ Hz}, 100 \text{ pulses.} \)

The ablation depth strongly depends on the local structure of the wood. The ablation rate is higher for the thin-walled spring wood cells and lower for the thick-walled late wood cells. Only a change of the wood colour produced by beginning carbonisation is found below the threshold. For low laser fluence the energy per volume in a thin sheet at the wood surface is too small for ablation. Therefore, the high surface temperature gives only rise to carbonisation. The same behaviour is observed for high power densities and high repetition rates. In this case, there is not enough time to cool down the surface between pulses because of the low thermal conductivity of wood.

Lowest ablation depth per pulse was observed for cross cuts, as expected from the anisotropy of the cellular structure. On the cross cut a large part of the incident power gets lost in the deep interior of the tube-like cells without effect. In the case of the other orientations, the effective thickness is much smaller, essentially by a factor given by the ratio of cell wall thickness to void diameter. This anisotropy is more pronounced for coniferous wood. The ablation rate strongly depends on porosity of the wood. Therefore, for beech, ablation rates were found about 50 % lower than for pine.

Changing from 308 nm to 10.6 \( \mu \text{m} \) with five-fold decrease of power density produces a 20-fold increase of ablation rate for pine (Figs. 7a,b). At low power densities, carbonisation and ablation have been observed. At higher power density, carbonisation is suppressed while the ablation rate is high.

Figure 7a. Ablation depth versus power density for pine (slash cut) irradiated by excimer laser. Parameters: \( \lambda = 308 \text{ nm}, T_p = 40 \text{ ns}, u = 19\%. \)

Figure 7b. Ablation depth versus power density for pine (slash cut) irradiated by CO2-TEA laser. Parameters: \( \lambda = 10.6 \mu \text{m}, T_p = 1.2 \mu \text{s}, u = 10\%. \)
About 20% higher ablation rates were found in UV for beech wood with 12% water content compared with that of 30%.

Apparently, an additional amount of pulse power is necessary for the evaporation of the additional water.

In-situ high speed photographs of the excimer laser ablation process of wood show that three main stages of the process can be distinguished from the visible part of the emission spectrum of the plume:

0...1 µs after laser pulse: absorption of the laser pulse and development of a thin light emitting plasma sheet on the surface,
1...200 µs after laser pulse: plasma expansion,
200 µs...1 ms after laser pulse: burning of gases coming out of the wood surface because of chemical processes caused by thermal effects.

The existence and intensity of the third stage strongly depends on repetition rate and power density. Distinct differences in the brightness were found for light emission of springwood and late wood indicating differential intensity of the ablation process.

At 308 nm and 10.6 µm, melting was observed for all components of the wood structure (for example Figs. 9a, 9b). The thickness of the molten zone is in the range of 1 µm. It means that also for laser wavelength in the UV range, ablation seems to be mainly caused by thermal processes, e.g. the excitation of phonons in the macromolecules of the wood components. The breaking of molecular bonds by direct interaction with a photon does not seem to be the dominating process. Ablation experiments on pure cellulose by parameters allowing an efficient ablation of wood ($\lambda = 308\ \text{nm}$, $E = 20 \cdot 10^6 \ \text{Wcm}^{-2}$) only causes a change of the cellulose colour accompanied by melting of a thin surface layer. Probably other wood components, such as lignin that glues the cell walls made of cellulose, effectively absorb the laser light. This seems to be supported by observation: even in the presence of molten and foam-like expanded cellulose from the cell walls, an open slit remains between the cells where the lignin had been (Fig. 9b).

Figure 8. High speed photographs of excimer laser ablation process on pine wood (quarter cut). Parameters: $\lambda = 308\ \text{nm}$, $T_p = 40\ \text{ns}$, $E = 20 \cdot 10^6\ \text{Wcm}^{-2}$.

Figure 9a. Melting of the surface during laser ablation of pine wood (cross cut) with CO$_2$-TEA laser is observed. Parameters: $\lambda = 10.6\ \text{µm}$, $T_p = 1.2\ \text{µs}$, $E = 4.3 \cdot 10^6\ \text{Wcm}^{-2}$, $f = 1\ \text{Hz}$, 30 pulses.

Figure 9b. Melting of the surface during laser ablation of beech (cross cut) for excimer laser is observed. Parameters: $\lambda = 308\ \text{nm}$, $T_p = 40\ \text{ns}$, $E = 20 \cdot 10^6\ \text{Wcm}^{-2}$, $f = 20\ \text{Hz}$, 100 pulses.
On laser ablated wood surfaces, improved penetration and therefore better adhesion for specially adapted adhesives and coatings is expected (figs. 10a,b) [7].

Figure 10a. SEM-image of sawn fir wood, cut parallel to the grain. No penetration of a PVAc-adhesive is shown. Viewable area is 250 µm.

Figure 10b. SEM-image of laser irradiated fir wood, cut parallel to the grain. Improved penetration of a PVAc-adhesive is observed. Viewable area is 200 µm.

3. PRELIMINARY EXPERIMENTS ON SEALING BY MELTING [8]

Carbonised or melted wood surface areas are well known but undesirable results of laser treatment of wood. They are usually removed. Thermal degradation is poorly controllable and unavoidable and as a result, the quality of machined wood will be reduced.

Parameswaran had discussed changes on wood during laser cutting and had given the following description: Brown and black coloured edges arise from the mainly thermal process of cutting and they are characteristic for combustion and carbonising of the cellular structure. As a result of melting and subsequent solidification, the with of the cells is reduced at the surface [9]. High temperature during the cutting process (about 700°C, Arai et al. 1979) results in conversion of the cellular structure to glassy solid substance. Back had calculated the melting temperature for cellulose as about 450 °C. Melting without combustion and carbonising should be possible if heating and cooling of the treated area are occurring in a very short time [10].

Figure 11. Pyrolysis temperature of wood constituents. [8]

Figure 12. Scheme of laser induced modifications on wood surfaces. The depth of laser treatment depends on wavelength λ, power density E, pulse duration Tp, and number of working steps.
Critical temperatures can be seen from Fig. 11 which shows the temperature of phase transitions such as melting and of pyrolysis for the main constituents of wood: cellulose, hemicellulose, lignin. The viscosity of lignin becomes apparent at about 100 °C. The pyrolysis temperature is lowest for hemicellulose: about 200 °C. For lignin, pyrolysis occurs at much higher temperature than softening, whereas for cellulose the two processes nearly coincide.

3.1 Experimental setup

For preliminary experiments, a CO₂-slab laser from Rofin-Sinar with 2.5 kW and pulse duration from 1 ms up to continuous mode was used. This type of laser was chosen because of the good absorption of wood in the range of 10 µm and the possible wide range of pulse duration. Small samples (20x20x20 mm) of beech and pine wood were used. Optical changes of the surface and depth of laser treatment while varying irradiation parameters (pulse duration/ repetition rate, average power and power density) were observed. The depth of laser treatment includes depth of ablation, depth of molten layer and depth of thermal/structural modifications except of melting.

3.2 Results of laser treatment and SEM investigation

With CO₂-laser and pulse width in the range of 1 ms, the melting of wood surfaces without carbonising is observed (Fig. 13). The depth of the molten layer was measured using light and electron microscopy. It was found to be in the range from 2 µm up to 7 µm. Therefore the depth of thermal/structural modifications was measured in the range from 5 µm up to 15 µm.

The surface temperature is assumed to be not higher than 200 °C because above this temperature, according to the known thermal behaviour of the wood components, carbonising of hemicellulose and lignin is expected.

![Figure 13. SEM-Image of laser irradiated pine wood, cross cut, CO₂-slab laser, Tp= 1 ms, melting of wood surface is observed.](image1)

![Figure 14. Change of moisture absorption for laser irradiated wood surface, pine wood, cross cut.](image2)

Changes in physical properties, i.e. moisture absorption of the surface are realized measuring the welling behaviour. For measurements, we have used a standard device for measuring the contact angle of a sessile drop. The surface tension of the liquid (distilled water) is well known. Short time measurements have shown that the moisture absorption slowed down remarkably.
4. USE OF LASER CLEANING IN PRESERVATION OF ARTWORKS [11,12,13]

The small Saxon town of Pirna near Dresden is famous for its historical center that has been thoroughly reconstructed and restored since 1991. One of the most remarkable buildings of this area of cultural heritage is the so-called Tetzelhaus where Johann Tetzel, the notorious peddler of indulgences, was born in 1465.

A wooden panel chamber was detected during preparatory investigations of the house before conservation and restoration, carried out by architecture office Milde & Möser. That chamber is an enclosed room with wooden plank ceiling and walls. The remains of narrow Gothic windows indicated an appreciable age, which has been confirmed by dendrochronology.

After a careful removal of the fake ceiling, the fir wood wall panels with dendrochronological age of 600 years turned out to be heavily damaged in some areas. The extent of damage was hard to judge because of dirt and multiple layers of tapestry and paint covering the wooden surface.

4.1 Reasons for the application of laser cleaning

Because of the poor state of the wood in the damaged parts, abrasive-cleaning techniques, such as dry blasting, had to be excluded. For finding a concept for a suitable cleaning procedure, the layers were tentatively removed mechanically with a scalpel, and with water vapour.

Applying the scalpel as a proven and well-established means turned out to be of low efficiency. In addition, it would have involved the danger of removing parts of the remaining wood together with the layers, especially in the seriously damaged parts of the panels (Fig. 16a). Applying water vapour results in higher efficiency but would have led to diffusion of various substances into the wood, resulting in grey stains (Fig. 16b) and eventually in difficulties with vermin extermination and consolidation of the substance. Considering these facts, a non-contact technique seemed to be more promising.

First tentative cleaning experiments with the cleaning laser quickly led to strikingly good results, judging from visual impression. The layers came off easily. Remains could be easily removed from holes and depressions without any visible damage to the wood (Fig. 16c).
Apparently, laser cleaning of large areas of wood of historical significance had never been tried before. There was no experience with laser cleaning of wood: no available data, not even for similar processes.

4.2 Proving harmless

Judging from laser cleaning of metals and stones, one should try to find out first whether wood cleaning is a selective and self-limiting process (which would ease the application). Also it must be ascertained that wood as a fragile organic material prone to thermal decomposition would not be affected by that part of the laser beam energy which turns into heat while being absorbed by the layer material.

The subjects of investigation were: mechanism of ablation, surface temperature of the wood, irradiation parameters for damage-free ablation, and the potential health hazard involved in the cleaning process.

4.3 Removal of layers on wood as a self-limiting process

The significance of the optical properties of the materials is explained in Fig. 17, where absorptivity is plotted over wavelength. It is seen that the absorptivity of beech and pine wood is minimal in the region around the wavelength of the Nd:YAG laser, $\lambda = 1064 \text{ nm}$, and much lower than that of graphite and Bohemian green soil, which have been selected as examples for coating constituents. It is also visible in Fig. 17 that the beam of Excimer and CO$_2$ lasers does not work very selectively on the chosen substances, because their absorptivities do not differ much. The significant difference between low absorptivity of wood and the higher absorptivity of the coating in the region of Nd:YAG laser wavelength is a precondition for a self-limiting process allowing the removal of the layer without harming the wood beneath.
The difference in absorptivity is also apparent through different plasma formations during ablation. Whereas with an power density of $283\times10^6$ Wcm$^{-2}$ per pulse, the irradiated coating forms a plasma plume, no such plume is formed during irradiation of the cleaned wood. The development of the plasma plume due to laser irradiation was recorded by a high-speed camera. It is obvious that about 5 µs after the end of the 6 ns pulse, the visible plume is still attached to the surface. This means that the plasma exists longer than the laser pulse does. The prolonged life of the plume near the surface increases the heating effect. Therefore, it is important to ensure that the temperature of the wooden surface stays below a critical value.

Consequently, one has to find those laser pulse parameters which guarantee a surface temperature below 100 °C during irradiation of clean wood. This set of parameters serves as an intensity threshold for wood cleaning. Irradiation with $283\times10^6$ Wcm$^{-2}$ pulses on a previously cleaned panel, with repetition and feed rate as usual, causes a pyrometrically measurable surface temperature of 73 °C (Fig. 18).

Therefore, this set of parameters can be considered safe for wood cleaning as the ablation threshold for the layers to be removed is lower, in some cases considerably lower. The parameter range for the self-limiting and safe cleaning of the coated panels was found to be $183\times10^6$ Wcm$^{-2}$ … $283\times10^6$ Wcm$^{-2}$.

The absence of any damage is demonstrated by SEM-pictures of cross-sections. Figs. 19a, 19b, 19c show sections of springwood and late wood before and after laser removal of the coating. It is obvious that even the thin layer (up to 60 µm) consisting of squeezed and torn cells and cell walls because of the original planing of the wood surface is still there. Thus, it can be stated that the process does not involve any "laser planing". This is convincing evidence of the self-limiting character of the process. These experiments led to the conclusion that ablation by shock waves as discussed by several authors does not seem to be relevant.

5. CONCLUSIONS

The study reports on fundamental aspects of these phenomena and relates different parameters such as wavelength $\lambda$, power density $E$, pulse duration $T_p$ and focus to the surface morphology that presents ablation or melting processes that can be seen from microscopic observations. Laser ablation of wood without carbonisation of the surface is possible for certain parameter sets. UV and IR-pulse lasers are suitable for this purpose because of the absorption properties of wood. For these laser types, melting of the cellulose in the µm range was observed, which supports the assumption of mainly thermal ablation at laser wavelengths above 300 nm. With CO$_2$-laser and pulse width in the range of 1 ms, the melting of wood surfaces without carbonising is observed. The depth of the molten layer was found to be in the range from 2 µm up to 7 µm. Therefore the depth of thermal/ structural modifications was measured in the range from 5 µm up to 15 µm. Short time measurements have shown that the moisture absorption of the laser irradiated wood surface slowed down remarkable. As a result of the difference in absorption behaviour of the coating to be removed and the wood surface, a damage free and self-limiting uncovering of wood surfaces from dirt and paint layers is possible.
Until recently laser application in wood technology has been restricted to a few techniques of cutting, perforating and engraving of wood and wood products. In view of this state of art it is highly justified to apply contemporary methods of material science to wood for better understanding of its structure and diverse properties in order to modify them with the aim of extended use. Presently a joint fundamental research program of the University of Technology Dresden and the Fraunhofer Institut Material and Beam Technology IWS is realized.

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REFERENCES