

Article

# Experimental Evaluation of ND: YAG Laser Parameters and Sample Preparation Methods for Texturing Thin AISI 316L Steel Samples

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**Featured Application:** Laser surface modifications for biomaterials, heat exchangers, electronics, nanotechnology, etc.

**Abstract:** In mechanical and material engineering, the effect of laser texturing depends on many factors besides device specification, primarily the properties of the materials being processed, and, secondly, the preparation of the sample. Laser texturing of thin (<5 mm) samples is mostly performed utilizing short-pulse lasers, but depending on the power of the laser beam, the process can also be performed by using continuous operation lasers. When using a laser beam to modify the surface layer, special attention should be paid to the surface preparation process. Engraving a shiny metal surface can lead to laser beam dispersion and energy loss. Some materials require special preparation and surface darkening in order to be effectively engraved. In the case of engraving, maximizing the efficiency and repeatability of the process is the key to obtaining the desired properties. The aim of the conducted study was to establish satisfying parameters and a sample preparation method for texturing thin AISI 316L samples. Appropriately selected laser parameters added to proper sample preparation. The sanding, etching, and darkening of the surface layer improved the quality of the weld and eliminated problems such as deformation and spark formation that often occur with raw samples during the texturing process.

**Keywords:** 316L; laser surface modifications; surface preparation; ND: YAG; laser engraving



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## 1. Introduction

The term ‘laser’ stands for *Light Amplification by Stimulated Emission of Radiation*. It is a commonly known name for a device that generates or amplifies electromagnetic radiation in the visible or invisible light range based on stimulated emission [1,2]. Looking back in history, the basis of modern laser technologies is research by Albert Einstein and his works on the photoelectric effect, quantum mechanics, quantum structure of light, and the possibilities of stimulating the emission of coherent, monochromatic, electromagnetic radiation [3]. One of the first industrial uses of a laser took place in the 1970s and involved using a beam to cut sheets of steel. Later, lasers were gradually used to modernize welding processes: welding, surfacing, heat treatment, surface modification, ablation, and cutting [3–10]. Various types of laser processing mechanisms and their applications were described by Kumar et al. in their extensive review article [2].

The most important elements of laser construction are the active medium placed in an optical resonator with two parallel mirrors spaced at a given distance and a pumping system that provides the energy needed to excite the active medium and needed to create the necessary conditions for the active medium to amplify the light passing through it.

These mirrors can be either spherical or flat. One of them is almost completely reflective. The reflectance of the second one must be lower than the first as it will act as the output mirror through which the radiation leaves the resonance cavity [11,12].

The light generated in this way has a number of unique properties, including the following:

- The form of a laser beam—it can either spread over long distances or concentrate a small, focused beam;
- A narrow emission spectrum;
- A continuous or pulsed emission;
- Coherent radiation, usually polarized;
- Low beam divergence;
- Short, high-power pulses.

Lasers can be divided into many categories depending on power, method of operation, applications, etc. Lasers are also divided into categories according to the phase, state, or resource of the gain medium, such as a solid (e.g., solid Nd: YAG, Ti-Shapphire, etc.), liquid (e.g., dye laser), gas (e.g., He-Ne, CO<sub>2</sub>, etc.), and semiconductor (e.g., GaAs, InAs, etc.). The fiber laser, which is currently widely used in the manufacturing industry, is a type of solid laser as the active amplifying medium is an optical fiber doped with solid rare earth elements such as erbium, ytterbium, neodymium, dysprosium, praseodymium, thulium, and holmium to ensure light amplification. The wavelength of the emitted light is determined by the energy levels of the various gain media. Fiber lasers have a wide pulse operating range, and their average power output can be up to several kW. Fiber lasers enable efficient industrial processing such as steel welding, thick metal cutting, texturing for dyeing, and metal 3D printing, etc. [2,3]. In addition to industrial applications, laser devices are used in many fields, e.g., in medicine, geodesy, military technology, astronomy, etc. [5–8,11,13–18].

Scientists from Slovenia used an Nd: YAG laser on ultrathin AISI 316 samples in order to enhance boiling heat transfer [18]. Polish scientists often use lasers to modify the surface layers of different biomaterials used in medicine and bioengineering [19]. A good description of the texturing was provided by Pou et al., who also used laser surface texturing for bioengineering applications [10]. It is very rare for researchers to share a detailed description of sample preparation, but a few can be found. One of them was published by Obona et al., who described the importance of polishing copper samples before the laser treatment [20]. Extensive experimental research conducted by Tam, Cheng, and Man provides readers with profound knowledge of brass laser surfacing. The scientists mention polishing and etching the samples before the laser treatment [21]. However, they do not explain the effect or importance of sample preparation.

In mechanical and material engineering, the use of laser radiation depends on many factors, primarily the properties of the materials being processed. Lasers are widely used in industry for surface treatment, marking products by engraving, precise cutting, and welding. There are also many subcategories of these processes, such as welding, cutting, surfacing, alloying, soldering, and laser micromachining, which includes microdrilling, microsurfacing, microwelding, engraving, ablative cleaning, and surface texturing. An exemplary division of mechanisms for laser modification of the surface layer was proposed by Kumar et al. [2]. The use of lasers in industry allows for improvements in efficiency and production speed while maintaining high precision, high quality, and repeatability of the process. An additional advantage is a clean workstation and the lack of waste materials during the machining process [2,11,12].

Material engraving with a laser beam can be used to mark the product, obtain specific visual and decorative effects, or modify the surface layer of a given material. Laser ablation involves the removal of material through the process of evaporation of surface layer material. Laser ablation is most often performed with short-pulse lasers, but depending on the power of the laser beam, the process can also be performed using lasers for continuous operation [12]. Performing laser modification to the surface layer both changes the composition of the alloy and gives it different microstructural features, but only on the surface of



the material, which often results in improved resistance to corrosion and the formation of fatigue cracks [6,22].

When using a laser beam to modify the surface layer, special attention should be paid to the surface preparation process [7,20]. In the case of engraving, maximizing the efficiency and repeatability of the process is the key to obtaining the desired properties. Engraving a shiny metal surface can lead to laser beam dispersion and energy loss. Some materials require special preparation and surface darkening in order to be effectively engraved [18,23].

The purpose of the conducted research was to select a suitable method for modifying the surface layer of AISI 316L stainless steel. AISI 316 is the second most commonly used stainless steel grade in the industry. The 316L version is a low-carbon variant of the 316 alloy, which makes the 316L stainless steel suitable for welding, which is especially important in the case of laser processing [24–26].

Due to the lack of detail about sample preparation in the literature and very little data about laser parameters such as power, frequency, pulse application time or speed, it seems to be very difficult for young researchers to decide on good starting parameters. The authors also noticed a lack of failure test descriptions. Since it is commonly stated that “one develops success from failures” and “discouragement and failure are two of the surest stepping stones to success”, the authors find it important to share both successful aspects of the study along with the failure-full path that precedes success. However, constantly starting the experiment from scratch and repeating the trial-and-error method lead to an immense waste of materials and energy. This waste could be avoided by simply sharing the knowledge of best practices and those that are usually classified as proper.

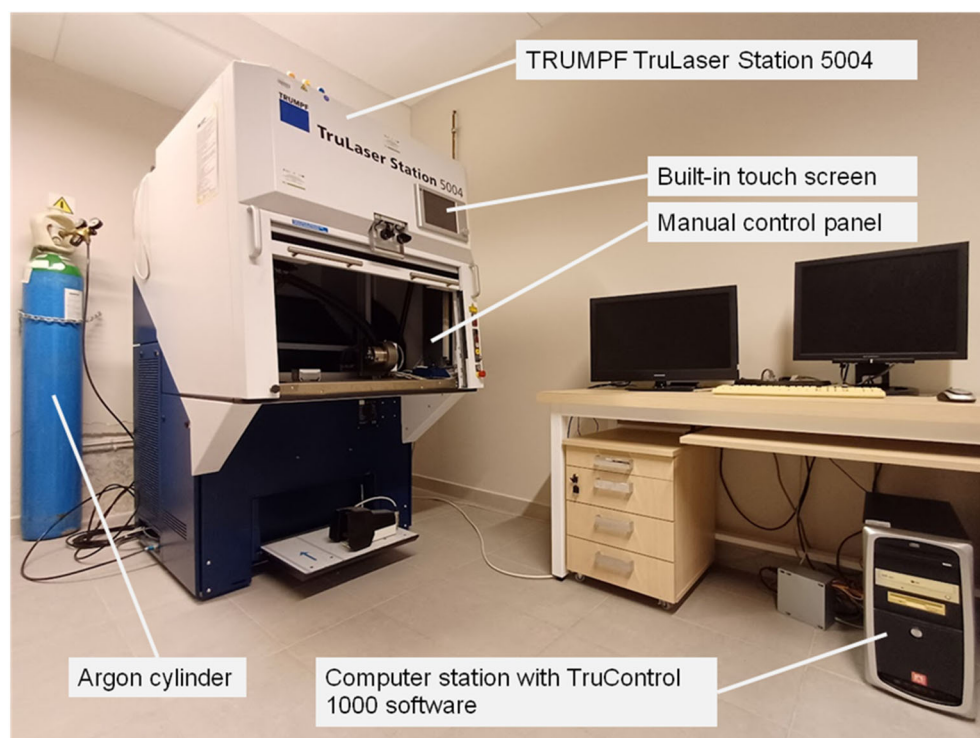
Thin (<5 mm) AISI 316L samples were subjected to laser texturing using different laser beam parameters. Then, they were experimentally scrutinized to see how much a proper sample preparation procedure affects the final result of the modification. The obtained effects were subjected to verification and microscopic observations, thus eliminating modifications that did not meet the objectives of the experiment.

## 2. Materials and Methods

The chosen method used to modify the surface layer of AISI 316L samples was the use of a pulsed Nd: YAG laser. The research station is located in one of the buildings belonging to the Faculty of Mechanical Engineering and Ship Technologies of Gdańsk University of Technology. The experimental setup presented in Figure 1 consisted of a TRUMPF TruLaser Station 5004, a German-made device with a built-in touch screen and manual control panel, and an adjacent computer station with dedicated TruControl 1000 software, version 1.92.0, provided by the manufacturer. At the back of the station, there was a cylinder with dedicated shielding gas. For this device, Argon 4.8 compliant with the ISO 14175-I1-Ar standard [27] was used.

The station was also equipped with an additional camera which enabled viewing of the head positioning and the laser melting process on a separate monitor. TruControl 1000 software allows one to design a modification path by developing a CNC program based on a list of available command codes and then selecting laser processing parameters. The program allows one to change three key laser parameters (power, frequency, and focus) in the range shown in Table 1. The ‘focus’ parameter approximately corresponds to the diameter of the laser spot. Built-in safety systems prevent the setting of dangerous parameters that threaten the user’s safety or threaten to damage the station. In some cases, this may result in automatic changes to some parameters or a blockage of the laser beam, preventing the remelting process from starting [28].





**Figure 1.** Nd: YAG TRUMPF TruLaser Station 5004 experimental setup.

**Table 1.** Range of available parameter values for the TRUMPF TruLaser Station 5004 laser.

Parameter	Value Range	Unit
power	300–6000	Watt [W]
frequency	0.2–833	Hertz [Hz]
focus	0.4–1.85	Millimeter [mm]

Based on a review of literature reports [2,18,29–32], it was decided that for the needs of the research application, which in this case was to be tested as a heat conductor compartment in a further study, the best solution was to create a simple modification path and use one CNC program with various machining parameters. This solution gives constancy in the form of a given modification path, while the variables are three laser parameters—power, frequency, and focus. A simple pattern was chosen, which resulted in performing remelting in a straight line from left to right, then moving the laser beam up the sample by a given distance and performing straight line remelting again, this time to the left, and then looping the sequence as many times as the sample size allowed. Other constant parameters were the pulse application time, which was 1 ms, and the speed of the laser head, which was set to 2 m/s as a default.

### 3. Results

Using the dedicated CNC program, a series of laser modifications of AISI 316L steel samples with dimensions of  $10 \times 10 \times 2$  mm was carried out. A simple pattern was chosen in order to focus on the parameters and sample preparation and not on coding. The laser beam went as follows: straight line from left to right (10 mm), then moved up (0.5 mm), a straight line again, right to left (10 mm), and then moved up (0.5 mm). The movement sequence was looped as many times as the sample size allowed. In the case of  $10 \times 10 \times 2$  mm samples, the size allowed for up to 10 sequences. The laser parameters were selected and changed based on current observations at the time. The constant parameters were the pulse time, which was 1 ms, and the laser head speed, which was set to 2 m/s.

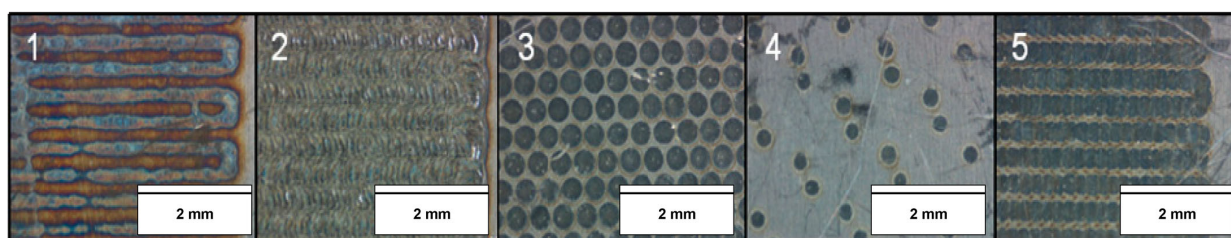
The program code for the first modification path series carried out on AISI 316L samples can be found in Supplementary Materials, Table S1.

The G0 H4 command starts the supply of shielding gas, where G04 T200 is responsible for the 200 ms waiting time before moving to the next step, and then G0 H3 starts the program. G91 is the command used to initiate the movement and application of the laser beam. The following part of the code is responsible for establishing and looping the movement commands. The software uses the relative coordinates written in relation to the axis of the X, Y coordinate system. Local X and Y coordinates were created by manually determining the starting point within the working space of the station, which was achieved by controlling the laser head positioning using the hand manipulator.

Conducting a series of modifications allowed the researchers to confirm the effective operation of the laser beam on AISI 316L steel without additional processing. By comparing the given laser parameters described in Table 2 with Figure 2, the visual effect of the modification of the surface layer is observed. Paths no. 1, 2, and 5 correspond to the shape of the programmed path with high accuracy, and individual pulses overlap, combining individual pulses almost into one line. Paths 3 and 4 were created after applying a lower pulse frequency, which resulted in their dispersion in the selected area.

**Table 2.** Laser processing parameters during the first series of modifications performed on AISI 316L steel.

Track No.	Power [W]	Frequency [Hz]	Focus [mm]	Observations:
1.	400	5.0	0.4	local burns, irregular edge of the pattern, sparkle formation, single dimples hard to identify, even spacing between dimples, dimples overlap;
2.	400	8.0	0.4	no local burns, irregular edge of the pattern, sparkle formation, single dimples not hard to identify, dimples strongly overlap;
3.	498	3.8	0.5	no local burns, regular edge of the pattern, less sparkle formation, single dimples easy to identify, even spacing between dimples, dimples do not overlap;
4.	400	1.0	0.5	some local burns, regular edge of the pattern, less sparkle formation, single dimples easy to identify, uneven spacing between dimples, dimples do not overlap;
5.	450	24.0	0.5	some local burns, regular edge of the pattern, sparkle formation, single dimples easy to identify, even spacing between dimples, dimples overlap.



**Figure 2.** Paths obtained by laser surface layer modifications performed on AISI 316L steel.

Laser power values entered manually were usually rounded to the nearest ten and frequency values to the nearest one; however, in the case of track 3, the security system made the correction itself. The obtained effect turned out to be so interesting that we decided to test the same parameters on target samples.

Visual and microscopic observations showed differences between samples with a surface layer modified with a pulsed Nd: YAG laser. Differences between individual remelts were observed due to differences in the given laser parameters. At this stage,



no significant defects were observed that would definitively disqualify the samples from further stages of testing. The resulting modification paths are repeatable.

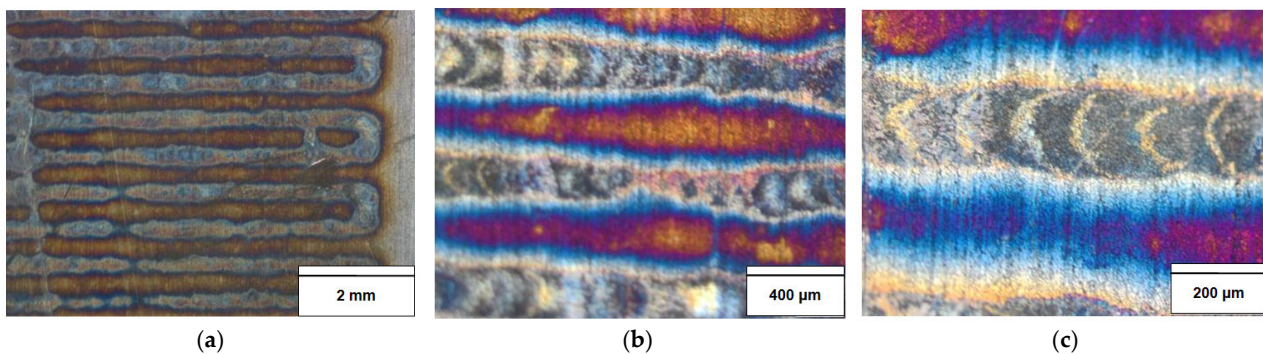
Recently, the CNC program has been modified for a target sample size of  $50 \times 50 \times 2$  mm. The second series paths covered a surface of  $48 \times 50$  mm with a tolerance of  $\pm 2$  mm depending on the positioning accuracy of the sample. With the larger modification area, the processing time increased to approximately 10 min per sample. In order to conduct effective texturing, the focus was increased to 1.5 mm and the power was increased to the range of 700–3000 W. The program code for the increased modification area can be found in Supplementary Materials, Table S2.

#### 4. Discussion

The simple CNC modification program designed by the authors covered a surface of  $10 \times 10$  mm with a tolerance of  $\pm 1$  mm depending on the accuracy of the sample positioning. The TRUMPF TruLaser Station 5004 used for the experiment did not have automatic sample positioning systems or automatic positioning of the laser head in relation to an object located in the working area of the device. Hence, minor inaccuracies in sample positioning relative to the point (0,0) of the coordinate system might have occurred. However, in the case of this experiment, this should not have a significant impact on the general results of the studies. The melting time was not precisely measured, but approximately ranged from several seconds to one minute.

Samples prepared using an Nd: YAG laser were observed on two light microscopes. A ZEISS Stemi 2000-C model was used to obtain a  $10\times$  magnification image, while the OLYMPUS VC50 BX51 model made it possible to obtain an image of the modifications made at  $50\times$  and  $100\times$  magnification. However, the  $100\times$  magnification allowed a focus only on selected details at a given depth of remelting, and usually did not provide a clear image of the entire dimple.

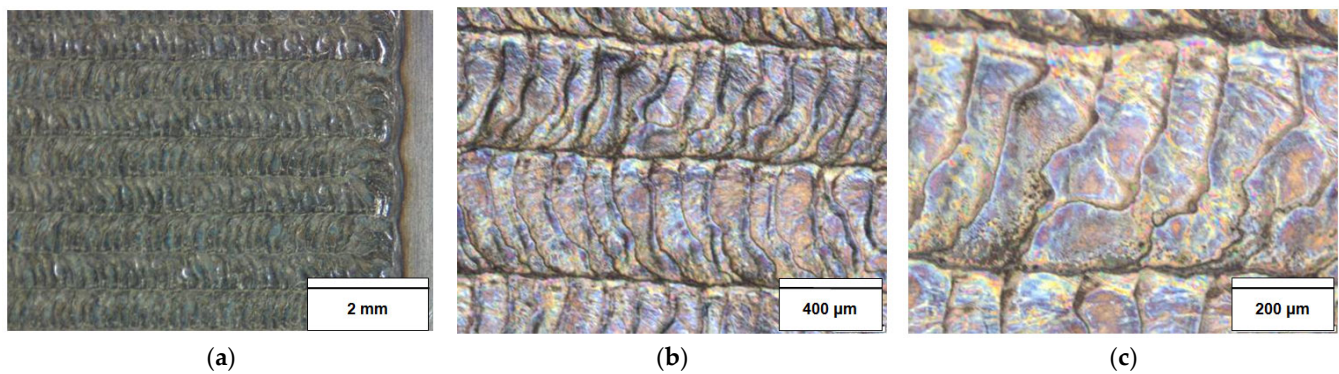
The first sample, presented in Figure 3, was melted in an irregular manner. The general shape of the modification corresponds with the movement commands, but the edges of the pattern are not consistent, and the size of a single dimple varies. Observations made using  $50\times$  and  $100\times$  magnifications show that individual dimples overlap, which is consistent with the experiment expectations. The single dimple diameter size is estimated to be  $200 \mu\text{m}$ . The direction of overlap alternates in rows in accordance with the direction of the movement of the laser beam. Individual rows of modification paths are spaced about  $400 \mu\text{m}$  apart. Many local burns were noticed, along with a strong sparkle formation during the process, which generates a risk of losing anti-corrosion properties. It also means that the procedure either contains a harmful combination of laser parameters or is lacking in base material protection. The path is also not repeatable, which means it cannot be taken into consideration for further tests and studies.



**Figure 3.** Microscopic observations of sample no. 1: (a)  $10\times$  magnification by ZEISS; (b)  $50\times$  magnification by Olympus; and (c)  $100\times$  magnification by Olympus.

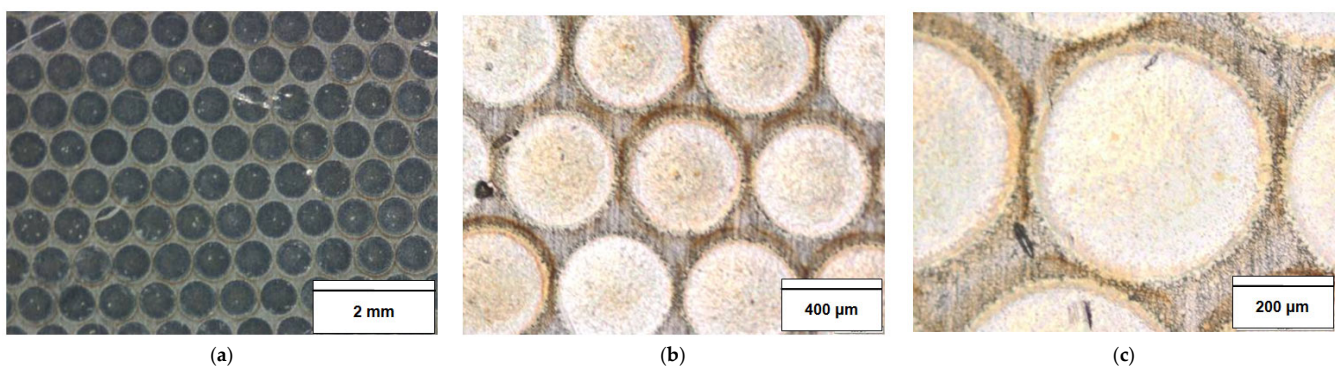
The second sample, presented in Figure 4, was also melted in an irregular manner. The general shape of the modification corresponds with the movement commands, but the

edges of the pattern are not consistent, and the sizes of the single dimples vary. Observations made using  $10\times$  magnification show that individual dimples strongly overlap. Moreover,  $50\times$  and  $100\times$  magnifications show that the ejected material almost completely covers the shape of the individual dimples. The direction of overlap alternates in rows in accordance with the direction of the movement of the laser beam. Individual rows of modification paths are not spaced apart. No local burns were noticed, although they might have been covered under the layer of ejected material. Some sparkles occurred during the process, but the damage was not as noticeable as it was on the first sample. This generates a risk of hidden spots with low mechanical and corrosion properties, which might result in a higher risk of cracking. It cannot yet be determined whether this procedure could be used for industrial purposes. Even though the individual meltings are not identical, the overall path seems to be repeatable.



**Figure 4.** Microscopic observations of sample no. 2: (a)  $10\times$  magnification by ZEISS; (b)  $50\times$  magnification by Olympus; and (c)  $100\times$  magnification by Olympus.

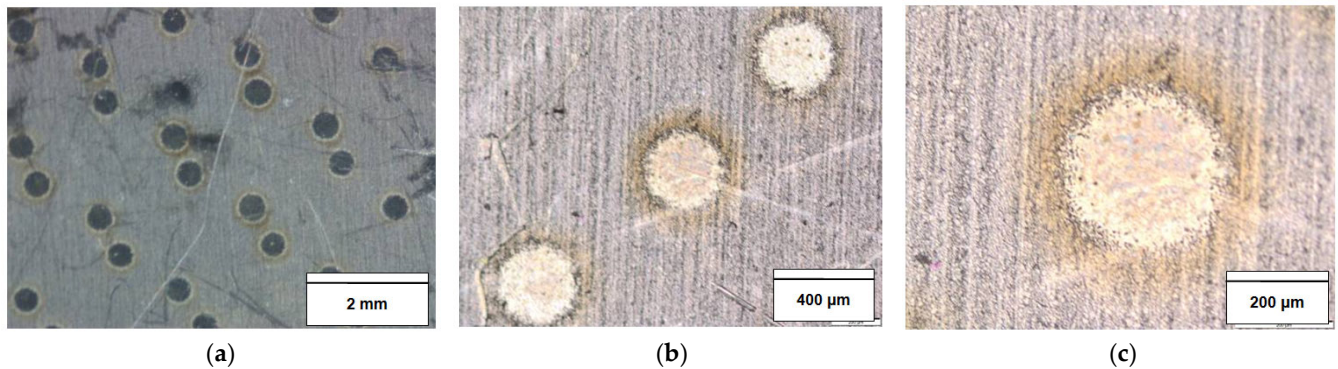
The most regular melting edges were formed on the third sample, presented in Figure 5. Compared to the other samples, the edges of the melting form an almost perfect circle, void of noticeable irregular edges. The combination of frequencies 3.8 Hz and 498 W power resulted in a regular honeycomb-like pattern. The focus was increased to 0.5 mm, which resulted in a larger-sized individual dimple. The approximate diameter of a single dimple is 0.48 mm. Dimples are spread evenly and do not overlap. The distance between the edges of the dimples is less than 0.05 mm. No local burns were noticed during the texturing process. Some local damage caused by sparkles can be noticed on the surface layer. One of them is clearly visible in Figure 5b. The texturing path is repeatable.



**Figure 5.** Microscopic observations of sample no. 3: (a)  $10\times$  magnification by ZEISS; (b)  $50\times$  magnification by Olympus; and (c)  $100\times$  magnification by Olympus.

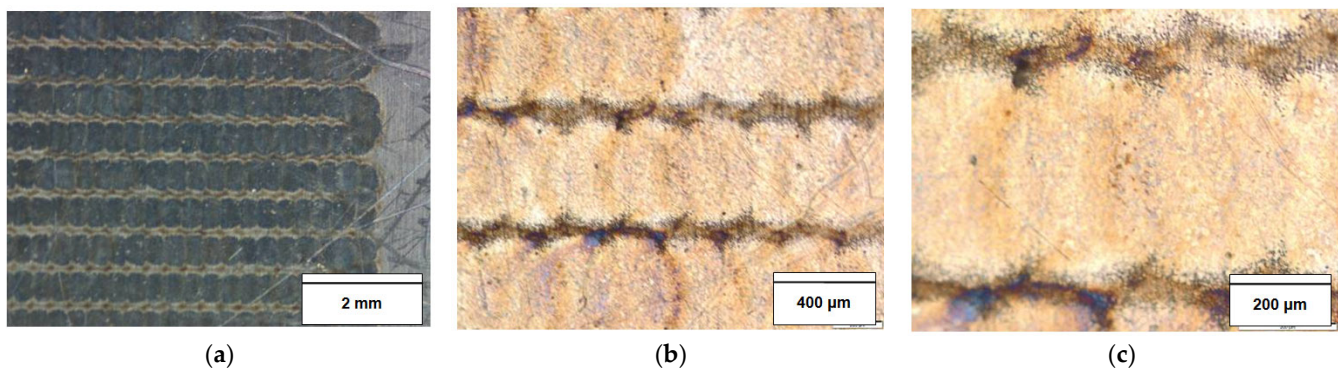
Sample no. 4, presented in Figure 6, was also melted in a regular manner and the individual dimples did not overlap. Even though the focus was set to 0.5 mm, the final diameter was no more than 0.3 mm. The distance between individual meltings was much

larger when compared to others. They varied from 0.5 mm to almost 2 mm. The path looked regular. It was repeatable up to a point where some distraction occurred, after which there was a path shift. The cause of the shifting remains unknown. It was assumed that there could have been some vibration in the building caused by a passing truck or some experiments conducted in an adjacent laboratory.



**Figure 6.** Microscopic observations of sample no. 4: (a) 10× magnification by ZEISS; (b) 50× magnification by Olympus; (c) 100× magnification by Olympus.

The fifth sample, presented in Figure 7, was created with increased frequency. Microscopic observations revealed some local burns which were not as intense as those in the first sample. A clear higher magnification image of this sample proved difficult to obtain. The estimated size of an individual dimple is 0.4 mm. The dimples overlap with a regular patterned edge. Sparkle formation caused visible damage to some areas of the sample.



**Figure 7.** Microscopic observations of sample no. 5: (a) 10× magnification by ZEISS; (b) 50× magnification by Olympus; and (c) 100× magnification by Olympus.

After increasing the laser power and the modification area it was observed that the lack of sample preparation produced some problems, such as

- Increased spark formation, which led to the destruction of the already applied path;
- Irregular shape of individual dimples;
- Local material burn-throughs;
- Deformation of the sample and upward bending during the remelting process caused by heating with the laser beam;

The observed undesirable side effects of the laser melting process were a motivating factor to take action to prepare the sample surface before exposing it to the laser beam.

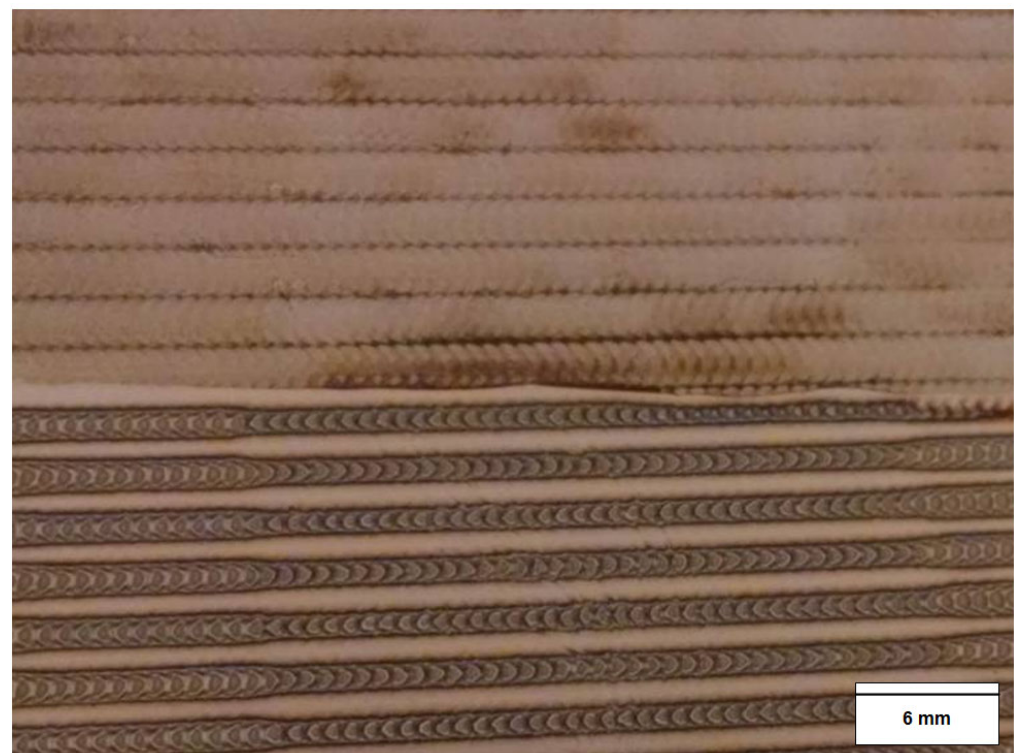
The sample preparation is an important, although rarely described in detail process. The literature data provide young scientists with only general statements, such as ‘polished’, ‘cleaned’ or ‘etched’, without a detailed description of the applied sample preparation methods. Vincenc Oboňa et al. mention that their copper samples were polished [20]. Pou



et al. used ultrasonical cleaner, ethanol, acetone, and distilled water both before and after laser treatment [10]. On the other hand, Kumar et al., in their literature review [2], claimed that effective laser ablation can be applied directly to the surface, without any need for additional pre-treatment, chemicals, or masking layers, which they take as an additional merit of reducing the overall process steps.

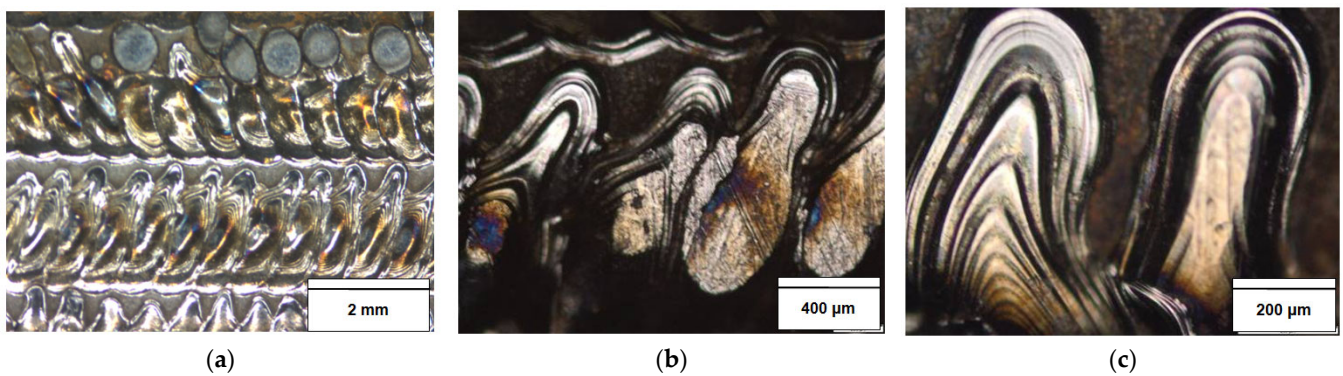
Attempts were made to grind, etch, and darken the sample surface. Standard sample preparation procedures were used for macroscopic tests based on the 'Metaloznawstwo: materials for laboratory exercises' script commonly used at the Gdańsk University of Technology [24,25]. The best effect was achieved by sanding the sample with 800-grit sandpaper using a manual two-disc grinder and polisher—Saphir 330, ATM GmbH (Blieskastel, Germany), a German product—and then etching with a mixture of concentrated (65%) hydrochloric and nitric acid in a volume ratio of 3:1, and subsequently darkening the sample surface with graphite or alcohol-based black ink from a common permanent marker. The sample prepared in this way was subject to much less deformation during the laser texturing process. With appropriately selected laser parameters, the problem of deformation was almost completely eliminated. Darkening the surface also prevented spark formation.

The visual difference between the remelting effect of the prepared and untreated surfaces can be observed in Figure 8, where only the upper part of the sample was ground, etched, and blackened, while the lower part remained unchanged. The figure shows how the final effect of applying the laser beam on the surface layer of the sample changes during a single sequence without changing the other parameters. The remelting process for this sample started from the lower left corner and ended in the upper right corner. Laser parameters were set as follows: power 2500 W, frequency 3 Hz, and focus 1.5 mm.

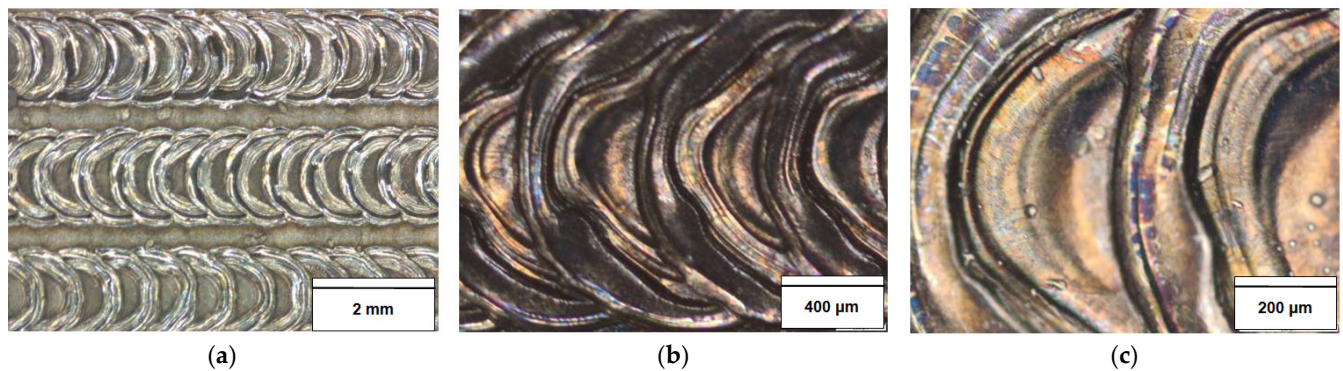


**Figure 8.** Comparison of the texturing effect of the etched and blackened surface (upper part of the sample) and the untreated surface (lower part of the sample).

Microscopic observation confirmed that the surface preparation makes a crucial difference in dimple formation. Figure 9 shows pictures of the raw AISI 316L surface subjected to laser texturing, while Figure 10 presents magnifications of the ground, etched, and darkened half of the same sample.



**Figure 9.** Untreated AISI 316L surface subjected to laser texturing of 2500 W laser beam power with pulse focus of 0.5 mm and frequency of 3 Hz: (a) 10× magnification by ZEISS; (b) 50× magnification by Olympus; and (c) 100× magnification by Olympus.



**Figure 10.** Etched and blackened AISI 316L surface subjected to laser texturing of 2500 W laser beam power with pulse focus of 0.5 mm and frequency of 3 Hz: (a) 10× magnification by ZEISS; (b) 50× magnification by Olympus; and (c) 100× magnification by Olympus.

The untreated steel surface subjected to laser beam modifications developed stretched, overlapping dimples with funnel-like shapes of material ejected during the ablation. Funnel-like, shaped dimple edges all followed the blowing direction of the shielding gas. It could be assumed that the stainless steel was melted, pushed out of the melt zone created by the laser beam, and moved by a steady blast of blown argon. The specific shape of dimple edges must have an impact on many physical properties, especially heat exchange and liquid flow parameters which could formulate another interesting research topic for further study. However, since the shape is mostly irregular and not repeatable, this phenomena cannot be taken into consideration as a modification technique. Also, strong pattern damage caused by sparkle formation can be observed, especially in Figure 9a,b. Some local burns can be observed at the bottom of each dimple.

On the other end of the sample, dimples created on the pre-treated surface look much more regular. The obtained pattern should be possible to recreate. No burns or sparkles were observed during the texturing process. Setting the focus parameter to 1.5 mm resulted in obtaining a dimple diameter of 1.48 mm. Dimples overlap evenly. Similar shapes of the dimples were obtained by Pou et al. [10], who also used Nd:YAG laser and compared its effectiveness in titanium texturing to a CO<sub>2</sub> laser. Scientists concluded that the processing assisted by the Nd:YAG laser was more suitable to modify the surface layer of titanium samples. They also found out that less splashes and more regular features were obtained using Nd:YAG rather than a CO<sub>2</sub> laser. However, the researchers emphasized that more research and further studies were needed.

Samples prepared this way could be taken into consideration for further study.

More analysis and measurements, including surface roughness tests, will be performed in the future. Since it was already discovered and described in many literature positions,

including the research conducted by Pou et al. [10], that laser treatment does not modify the chemistry of the samples, metallographic examinations were deemed unnecessary.

The samples must stay under unchanged conditions due to planned thermodynamical experiments. So far, the researchers were obliged to avoid destructive testing until the thermodynamical stage of an ongoing PhD study [28,33,34] was finished.

## 5. Conclusions

As for the further stages of the study, the following conclusions have been formulated:

1. The best texturing effect was obtained by a power range of 400–500 W applied to  $10 \times 10 \times 2$  and 1800–2500 W applied to  $50 \times 50 \times 2$  samples;
2. Samples previously treated with 800-grit sandpaper, then etched in a mixture of concentrated (65%) hydrochloric and nitric acid, and then darkened with graphite or black alcohol-based ink were subject to much less damage and deformation during the laser process;
3. The proper selection of laser parameters almost completely eliminates the problem of sample deformation;
4. Surface darkening successfully prevents spark formation.

At a later stage of the experiment, several additional changes will be introduced to the code. This should allow for better representation of the desired shape of the path with different laser beam parameters, without the need to create an individual code for each sample. This is in accordance with the previous assumption of adopting the modification path as a constant for the modification process.

**Supplementary Materials:** The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/app132212352/s1>, Table S1. The program code for the first modification path series carried out on AISI 316L samples; Table S2. The program code for the increased modification area.

**Author Contributions:** Conceptualization, E.K.; methodology, E.K.; software, E.K.; validation, S.G.-Z.; formal analysis, E.K. and S.G.-Z.; investigation, E.K.; resources, E.K.; data curation, E.K.; writing—original draft preparation, E.K.; writing—review and editing, S.G.-Z.; visualization, E.K.; supervision, S.G.-Z.; project administration, E.K. and S.G.-Z. All authors have read and agreed to the published version of the manuscript.

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