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Valentin Lang, Andreas Rank, Andrés F. Lasagni

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Direct laser interference patterning of metallic sleeves for roll-to-roll hot embossing

Valentin Lang^{*a,b}, Andreas Rank^b, Andrés F. Lasagni^{a,b}

^aFraunhofer-Institut für Werkstoff- und Strahltechnik IWS, Winterbergstr. 28, 01277 Dresden, Germany; ^bTechnische Universität Dresden, Institut für Fertigungstechnik, George-Bähr-Str. 3c, 01062 Dresden, Germany

ABSTRACT

Surfaces equipped with periodic patterns with feature sizes in the micrometer, submicrometer and nanometer range present outstanding surface properties. Many of these surfaces can be found on different plants and animals. However, there are few methods capable to produce such patterns in a one-step process on relevant technological materials. Direct laser interference patterning (DLIP) provides both high resolution as well as high throughput. Recently, fabrication rates up to $1 \text{ m}^2 \cdot \text{min}^{-1}$ could be achieved. However, resolution was limited to a few micrometers due to typical thermal effects that arise when nanosecond pulsed laser systems are used. Therefore, this study introduces an alternative to ns-DLIP for the fabrication of multi-scaled micrometer and submicrometer structures on nickel surfaces using picosecond pulses (10 ps at a wavelength of 1064 nm). Due to the nature of the interaction process of the metallic surfaces with the ultrashort laser pulses, it was not only possible to directly transfer the shape of the interference pattern intensity distribution to the material (with spatial periods ranging from $1.5 \text{ } \mu\text{m}$ to $5.7 \text{ } \mu\text{m}$), but also to selectively obtain laser induced periodic surface structures with feature sizes in the submicrometer and nanometer range. Finally, the structured nickel sleeves are utilized in a roll-to-roll hot embossing unit for structuring of polymer foils. Processing speeds up to $25 \text{ m} \cdot \text{min}^{-1}$ are reported.

Keywords: direct laser interference patterning, laser induced periodic surface structures, high-speed fabrication, roll-to-roll processing, surface functionalization

1 INTRODUCTION

Bionics provides an inexhaustible source of scientific inspiration [1]. Nowadays, several efforts have been devoted to understand the relationship between the surface topography/chemistry and the surface function. One famous example is the Lotus Effect, where micrometer and nanoscaled surface features enable hydrophobic and anti-adhesive properties [2, 3]. Other studies investigated the ability of periodic patterns to control the tribological performance (i.e. friction and wear reduction) or reducing drag by mimicking the structure of a shark's skin [4–10]. Moreover, surfaces presenting nanostructures have shown to be capable of optimizing the optical properties of surfaces (i.e. for light trapping) [11]. Another example is the skin of the Springtails (also called Collembola), which has a combination of submicrometer features arranged in hexagons, showing bacteria repellent properties. Furthermore, hierarchical structures (or multi-scaled) have shown to provide surfaces with hydrophobic and antiadhesive properties, simultaneously [12, 13].

Although, several of these examples have been exhaustively investigated, permitting to understand the mechanism involved for the improvement of the surface performance, just few of them can be used today in real products. This is mainly related to the complexity of existing fabrication methods, their high cost or their low throughput.

In addition to the fundamental investments (machines, infrastructure, etc.) that are required in every fabrication process, the speed of the method (throughput) is decisive. In [14], a comparison of different microstructuring fabrication methods was performed. Fabrication methods, such as conventional direct laser writing (DLW) or micro milling, are capable to run comparatively with high throughputs, but the feature that can be produced are generally larger than a few micrometers ($5 - 15 \text{ } \mu\text{m}$).

*valentin.lang@tu-dresden.de; phone +49 (0) 351 463 37844; fax +49 (0) 351 463 37755;
www.tu-dresden.de

Other methods, such as electron beam lithography (EBL) or laser interference lithography (LIL), provide feature sizes below 1 μm , but require the use of photoresists, which means that the structures cannot be directly produced in a one-step process. Furthermore, especially when producing surface textures with feature sizes in the nanometer range, extremely long processing times are required (several minutes up to some hours for 1 cm^2 of surface area).

An additional possibility for fabricating structures with feature sizes below 1 μm is the so-called laser induced periodic surface structures (LIPSS) method. While this phenomenon was first reported in the 1970s, in the last recent years new impulses in researching LIPSS have been arisen from the application of ultra-short pulsed lasers in the pico- and femtosecond regime [15, 16]. These periodic surface structures are generally smaller than the utilized laser wavelength, parallel or perpendicular to the laser beam polarization and have been observed after irradiating the material with a large number of laser pulses [15-18].

A technology capable to provide both, high resolution and throughput is direct laser interference patterning (DLIP). In addition, both the investment and the infrastructure required in DLIP are very similar to conventional laser writing, which means that neither vacuum nor clean room is needed.

DLIP enables processing of a wide variety of materials (polymers, metals and coatings) permitting to produce a large number of pattern geometries (e.g. line-, dot-, or cross-like pattern) in a one step-process [19–29]. Fabrication speeds up to approximately 1 $\text{m}^2 \cdot \text{min}^{-1}$ have been already reported, when using high-power ns-lasers [14]. In this method, an interference pattern is obtained by overlapping a certain amount of coherent laser beams. This is realized by splitting a main laser beam into several sub-beams using different optical configurations [30]. The periodic variation of the laser intensity described by the interference pattern is later transferred to the material's surface by selective laser ablation or modification processes at the positions corresponding to the interference maxima.

In the case of two-laser beams, a line-like periodic intensity distribution and its repetitive distance (pitch) can be controlled by varying the angle between the laser sub-beams. For this configuration, the spatial pitch (Λ) can be calculated as function of the laser wavelength (λ) and the mentioned angle (α) as follows:

$$\Lambda = \frac{\lambda}{2 \cdot \sin(\alpha/2)} \quad (1)$$

As it can be observed, for short laser wavelengths (i.e. for UV laser systems) resolutions even in the nanometer range are feasible ($\Lambda \sim \lambda/2$).

In this study, we focus on the fabrication of multi-scaled micrometer and submicrometer structures using the DLIP method with infrared picosecond laser pulses. The geometry and features produced are characterized using scanning electron beam and confocal microscopy methods. Finally, large area Ni-sleeves are textured and used in preliminary investigations of roll-to-roll hot embossing of PET foils. In this way, throughputs even one order of magnitude faster to the state of the art are reported.

2 MATERIALS AND METHODS

Direct laser interference patterning was used to create line-like patterns on the surface of nickel substrates. The experiments were divided into two sections. Firstly, ps-DLIP was performed on Ni planar substrates for investigating the effect of the laser processing parameters on the surface topography. After that, selected processing parameters were used for the processing of the cylindrical drum tools (sleeves).

In both cases, the substrates were translated by moving the workpiece relative to the laser beam using translational stages. Depending on the repetition rate used (number of laser pulses per unit of time) and the speed of the different stages, the pulse-to-pulse overlap was controlled. In addition, the laser fluence (ϕ , laser energy per unit of area) was varied.

For the processing of seamless nickel drum sleeves (Saechsische Walzengravur GmbH; diameter of 299.2 mm; thickness of 200 μm and a width of 300 mm) the setup shown in Figure 1 was used. The surface structures were produced by using a two-beam DLIP optical head (Fraunhofer IWS). In order to enable proper interference of the sub-beams on the substrate, focusing of the optical head is arranged by movement of a linear stage in Z direction (Aerotech ATS150-200).

For treatment of certain areas, the optical head is translated by an additional linear stage in X direction (Aerotech ABL80100) and the workpiece is moved relative to the optical assembly either by a rotary axis (Aerotech ATS150-200) or an optional linear stage in Y direction (Aerotech ABL15030). The utilized laser was Nd:YAG solid state laser (Edgewave PX200) emitting picosecond pulsed ($\tau = 10$ ps) near infrared radiation ($\lambda = 1064$ nm). Laser power measurements were executed using a power meter (Newport PMKIT-24-01). A rotatable half-wave-plate in combination with a polarizing beam splitter was used to control the laser output power. The experiments were carried out at ambient conditions [31].

Finally, a roll-to-roll hot embossing system (Coatema Basecoater BC 51) for the processing of PET foils was used. The system permits variation of the linear web speed of the foil from 1 to 50 m·min⁻¹.

Both confocal (Sensofar S NEOX) and scanning electron microscopy (Philips XL30 ESTEM FEG) were used to characterize the treated samples (both, Ni and PET substrates). Analysis of elementary values for the description of periodic surface patterns, such as structure height (ISO 5436-1), Power Spectral Density studies (ISO 10110-8) and isotropy calculation (ISO 25178-2) were performed.

Figure 1. DLIP - experimental setup: (a) photograph of the DLIP patterning system used for the processing of Ni-sleeves; (b) DLIP treated Ni drum sleeve with spatial pitch of $\Lambda = 2.8$ μm and (c) schematic drawing.

3 RESULTS AND DISCUSSION

As mentioned in the previous section, DLIP was used first for structuring Ni-planar substrates (using an IR ps-laser system). The laser fluence, pulse-to-pulse overlap and the spatial period of the interference patterns (Λ^1) were systematically varied. Examples of processed surfaces with spatial pitches (Λ^1) of 1.5 μm , 2.8 μm and 5.7 μm are shown in Figure 2. The images show that for all used spatial periods, well organized periodic surface patterns could be fabricated. However, in addition to the primary periodic structures, characterized by the line-like pattern (with spatial period Λ^1), two other repetitive patterns, denoted as Λ^2 and Λ^3 , have been observed. The latter two patterns, present length scales (repetitive distances Λ^2 and Λ^3) smaller than the used laser wavelength (1064 nm).

Figure 2. SEM images of DLIP-treated Nickel substrates: (a, c) $\Lambda^1 = 2.8 \mu\text{m}$, $F_p = 557 \text{ mJ}\cdot\text{cm}^{-2}$ (b, d) $\Lambda^1 = 2.8 \mu\text{m}$, $F_p = 116 \text{ mJ}\cdot\text{cm}^{-2}$ (e, g) $\Lambda^1 = 5.7 \mu\text{m}$, $F_p = 557 \text{ mJ}\cdot\text{cm}^{-2}$ (f, h) $\Lambda^1 = 1.5 \mu\text{m}$, $F_p = 557 \text{ mJ}\cdot\text{cm}^{-2}$.

For samples treated with high pulse-to-pulse overlaps, but low laser fluences (ϕ_p), both HSFL and LSFL are observed only at the interference maxima positions (see Figure 2b and d). Furthermore, the observed surface patterns are relative flat, which means that ablation at interference maxima positions is negligible. For higher laser fluences (i.e. Figure 2a and c), significant ablation of the metallic material is observed at the maxima positions and the LIPSS are also present at the minima [31].

An important parameter describing the topography of the produced surface patterns is the structure height h_{st} . Figure 3 shows confocal microscope images of selected treated Ni surfaces with spatial pitches of $\Lambda = 1.5 \mu\text{m}$ (Figure 3a and b) and $\Lambda = 2.8 \mu\text{m}$ (Figure 3c and d). For each spatial pitch, two cases are illustrated corresponding to pulse-to-pulse overlaps of 80 % and 95 %. The selected examples show two different behaviors depending on the spatial period of the interference pattern (Λ^1). For the $1.5 \mu\text{m}$ periodic structures, a higher pulse-to-pulse overlap (95 %, see Figure 3a) leads comparatively to periodic patterns with a low pattern quality (isotropy = 48.6 %: an isotropy value of 0 % indicates a perfect periodic line-like pattern while 100 % is obtained for a totally random surface). In contrast, patterns produced with a lower overlap (80 %, see Figure 3b) present better pattern quality, with isotropy values near to 0 % (0.75 %), denoting an almost perfect periodic line-like pattern.

Figure 3. Confocal microscope plots of DLIP-treated Nickel surfaces with: (a) $\Lambda^1 = 1.5 \mu\text{m}$, $F_p = 557 \text{ mJ}\cdot\text{cm}^{-2}$, pulse-to-pulse-overlap 95 %; (b) $\Lambda^1 = 1.5 \mu\text{m}$, $F_p = 557 \text{ mJ}\cdot\text{cm}^{-2}$, pulse-to-pulse-overlap 80 %; (c) $\Lambda^1 = 2.8 \mu\text{m}$, $F_p = 557 \text{ mJ}\cdot\text{cm}^{-2}$, pulse-to-pulse-overlap 95 %; (d) $\Lambda^1 = 2.8 \mu\text{m}$, $F_p = 557 \text{ mJ}\cdot\text{cm}^{-2}$, pulse-to-pulse-overlap 80 %.

On the contrary, as shown in Figure 3c and d, for comparatively larger spatial pitches ($\Lambda^1 = 2.8 \mu\text{m}$) the pattern quality is not distorted for large pulse-to-pulse overlaps. For the two shown examples, the pattern quality remains almost constant for the two used conditions. For the 80 % and 95 % pulse-to-pulse overlaps, the measured isotropy values were 1.10 % and 1.19 %, respectively [31].

A possible explanation for this behavior can be found in [29, 32]. In the mentioned studies, a correlation between the pattern quality and the maximal achievable structure depth was found to be dependent on the thermal diffusion length.

Thus, periodic patterns with short spatial periods generally show lower homogeneities as well as reduced structure heights.

The systematic investigation of the influence of the pattern pitch, laser fluence (φ) and pulse-to-pulse overlap on the structure height is shown in Figure 4. Three spatial pitches (1.5 μm , 2.8 μm and 5.7 μm) are compared. The results indicate that for all pitches an increasing pulse overlap leads to deeper structures (Figure 4a).

The images do not only describe the evolution of the structure height as function of the above mentioned parameters, but also the aspect ratio (Figure 4b). The latter is defined as the quotient between the structure height and the pitch of the line-like pattern. As it can be observed, the highest aspect ratio (~ 0.74 , structure height $\sim 2.1 \mu\text{m}$) is achieved for a spatial pitch of 2.8 μm at the maximum fluence ($\varphi = 557 \text{ mJ}\cdot\text{cm}^{-2}$) and pulse-overlap (95 %) [31].

Figure 4. (a) Structure height (h_{st}) and (b) aspect ratios of fabricated patterns as function of the pulse-to-pulse-overlap and laser fluence for spatial pitches of 1.5 μm , 2.8 μm and 5.7 μm .

In the case of the 1.5 μm spatial pitch, the maximal achieved structure height was $\sim 0.45 \mu\text{m}$ corresponding to an aspect ratio of 0.33. For the larger spatial pitch of 5.7 μm , the maximal achieved structure height was also approximately 2.1 μm , like for the 2.8 μm patterns. These results indicate that for spatial periods larger than 2.8 μm , saturation on the structure depth with increasing laser fluence and the pulse-to-pulse overlap can be observed. For shorter spatial periods,

such 1.5 μm , the maximal achievable structure height is significantly reduced and similarly as before. This effect can be explained by thermal diffusion processes [29, 32].

A major objective of DLIP treatment of nickel substrates was not only to produce multi-scaled surface patterns in a one-step process, but also to use them as stamping tools for a high throughput roll-to-roll hot embossing process. Hence, the obtained findings concerning the application of DLIP on planar Nickel substrates built the basis for the treatment of the cylindrical tool parts (sleeves).

Using the information depicted in Figure 4, a collection of selected parameters was applied for the treatment of the Nickel drum sleeves. This parameter set consisted of patterns produced with the three evaluated spatial pitches (1.5 μm , 2.8 μm and 5.7 μm) utilizing different irradiation conditions (pulse overlaps). In all cases, the laser fluence was set to $\phi_p = 0.363 \text{ J}\cdot\text{cm}^{-2}$. For the hot-embossing process, the temperature of the lower roll was set to 45° C while the roll with the imprinting sleeve was set to a temperature of 77° C. The liner speed of the foil was first set to 1 $\text{m}\cdot\text{min}^{-1}$ and the applied pressure was 6 bar.

Figure 5. Confocal (a, b) and scanning electron microscope (c, d) images of hot embossed PET foils with DLIP treated Ni-tools. (a, c) $\Lambda = 1.5 \mu\text{m}$; (b, d) $\Lambda = 2.8 \mu\text{m}$; Hot embossing temperature of lower drum: 45° C; temperature of structured drum: 77° C; linear speed: 1 $\text{m}\cdot\text{min}^{-1}$, pressure: 6 bar.

Figure 5 shows a brief selection of realized surface pattern on the PET foils. Structures produced on the PET films with spatial pitches of 1.5 μm and 2.8 μm are shown (Figure 5a, c and Figure 5b, d, respectively). In the case of the $\Lambda' = 2.8 \mu\text{m}$ structures (Figure 5b and d), the obtained periodic structure shows a structure height ($h_{st,PET}$) of 0.56 μm with an isotropy of 1.04 % (denoting a nearly perfect periodic line-like structure). In comparison, the structure height of

the Ni sleeve was $1.12\ \mu\text{m}$ with an isotropy of 1.33 %. In the case of the $1.5\ \mu\text{m}$ pitch, the realized pattern on the PET foil shows a structure height of approximately $0.16\ \mu\text{m}$ with an isotropy of 0.64 %. The structure height of the periodic pattern at the Ni tool for this condition was very similar ($0.17\ \mu\text{m}$ and an isotropy of 1.05 %).

In general, the obtained topography on the PET foils by the hot-embossing roll-to-roll process showed overall a lower structure height compared to the Ni-tool, especially for the large spatial periods. This fact can be explained by improper filling of the PET into the cavities of the Nickel tools. As side effect the topography of the embossed PET foils is also smoothed compared to the topography of the tools. This effect can be confirmed, since the LSFL can be observed in the processed PET foils but not the more filigree HSFL [31].

Finally, preliminarily experiments describing the influence of the foil linear-speed on the structure height of the embossed PET material was investigated. This dependence is depicted in Figure 6. The results show a decrease in the structure height of the embossed polymer, principally for the larger spatial pitches ($2.8\ \mu\text{m}$ and $5.7\ \mu\text{m}$). In contrast, the structure height of the embossed PET foil for a pitch $\Lambda' = 1.5\ \mu\text{m}$ keeps almost constant across the entire range of applied processing speeds ($1 - 25\ \text{m}\cdot\text{min}^{-1}$). However, the latter case accords to significantly smaller structure heights compared to the large pitches. In order to achieve constant structure heights with increasing processing speed, further investigations must be conducted with the aim of compensating this detected effect by fine-adjustment of the tool temperature and pressure.

Figure 6. Structure height (h_{st}) of produced surface patterns on the PET foils as function of the foil linear speed for $\Lambda = 1.5\ \mu\text{m}$ (pulse-to-pulse overlap = 60 %), $\Lambda = 2.8\ \mu\text{m}$ (pulse-to-pulse overlap = 90 %) and $\Lambda = 5.7\ \mu\text{m}$ (pulse-to-pulse overlap = 90 %).

4 CONCLUSIONS

Three level multi-scaled periodic patterns, with feature sizes in the micrometer and submicrometer range, could be fabricated on Ni surfaces using ps-DLIP. While the size of the primary structure could be defined by adjusting the angle between the interfering laser beams (from $\Lambda' = 1.5\ \mu\text{m}$ to $5.7\ \mu\text{m}$), low and high spatial frequency laser induced periodic surface structures, with pitches of approximately $\Lambda^2 = 800\ \text{nm}$ and $\Lambda^3 = 160\ \text{nm}$, respectively, were also observed. In addition, the dependence of both structure height and structure quality with the laser fluence and pulse-to-pulse overlap

were investigated. For all utilized spatial pitches (from 1.5 μm to 5.7 μm) a direct correlation between the structure height with the pulse laser fluence as well as the pulse-to-pulse overlap was observed. Finally, using a set of optimized processing parameters, large area Ni-drums were treated using the ps-DLIP method. These molds were later employed in preliminarily investigations of roll-to-roll hot-embossing of PET foils. In general, a reduction of the structure height of the imprinted periodic PET structures with increasing linear-speed was observed. However, this research demonstrates the potential of this method for high throughput surface functionalization.

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