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Precision structuring and functionalization of ceramics with ultra-short laser pulses

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High-performance ceramics have been firmly established for the manufacturing of tools and components in the modern electronics industry and mechatronics. Various components such as circuit boards, bearings, and sensors benefit from their specific characteristics, such as wear resistance, stiffness, and electrical neutrality. Apart from these advantages, the brittleness and hardness of ceramics turn the mechanical processing into a challenging and difficult task. Against this background, modern laser technologies have already been used to process ceramics for many years, enabling a contactless and wear-free machining. However, regarding high precision applications, for instance, the drilling of micro-holes or the fabrication of well-defined cavities and three-dimensional structures, conventional laser processes reach their limits. Especially due to thermal influences of the laser radiation, brittle edges, stresses, and redeposited layers emerge. Ultrashort pulse lasers enable completely new processing qualities in these fields. The extremely short pulse durations within the pico- and femtosecond range lead to nonlinear absorption mechanisms and an almost athermal material removal. Thereby, dielectric materials can be processed precisely and gently. In the course of a comprehensive process study, the beam–material interactions of ultrashort pulses with ceramics have been investigated. Besides the material properties, the ablation process is influenced by a multitude of laser parameters, such as wavelength, pulse overlap, and fluence. In order to reveal the most important variables, the experiments have been conducted by applying modern statistical methods. Using alumina (Al2O3) as an example, it is shown how different parameter regimes lead to disparate process qualities and efficiencies. The generated models have been used to optimize industrially interesting applications, from the separation of ceramic printed circuit boards to the realization of precise design structures. © 2018 Laser Institute of America. https://doi.org/10.2351/1.5040628

Key words: ultra-short pulses, ceramics, ablation, surface structuring, DoE

I. INTRODUCTION

The advantages of ultrashort pulses are well known. Since the plasma expansion and thermal diffusion are negligibly small, material damages such as stresses, melt, and debris can be significantly reduced, which is particularly suitable for the high precision processing of dielectric materials.1 On the other hand, ultrashort pulse lasers are often associated with small removal rates and the resulting poor economic efficiency of the processes. Accordingly, there is always the question of how the process velocity can be increased in order to make the ultrashort pulse technology interesting for industrial applications. The first simple step is often the optimization of the various influencing parameters. Based on a process analysis of the ultrashort pulse ablation of ceramics, it is shown how qualitative and quantitative aspects can be optimized to solve current industrial problems.

A. State of the art

As summarized by Samant and Dahotre, previous investigations concerning the laser processing of ceramics especially focused on cutting and drilling using CO2 and Nd:YAG lasers in particular.2 Despite the already widespread use of lasers in ceramic processing, there are still some challenges to be overcome, such as the avoidance of cracks and debris. Ultrashort pulse durations are especially suited to achieve high surface qualities, even with regard to brittle-hard, dielectric substrates. As shown by Radtke for the ablation of alumina, a decrease in the pulse duration from 150 to 10 ns can already reduce the thickness of redeposited layers and enable a partial removal of large ceramic grains.3 A further reduction of the pulse duration down to femtosecond pulses is advantageous to obtain micro-structures with a particularly high edge quality, which has been described by Perrie et al. concerning alumina.4 Kim et al.5 and Chen et al.6 give further information about threshold fluences, surface morphologies, and chemical changes referring to the one-dimensional single and multi-pulse processing of alumina and aluminum nitride.

However, regarding the three-dimensional surface ablation of ceramics, further investigations are necessary to determine significant parameter correlations as well as process
limits. Due to the heterogeneous microstructure of ceramics, the interactions between beam and material emerge from a combination of photothermal and -chemical processes. Depending on the laser parameters and the type of ceramic, a multitude of physical phenomena, such as melting, dissociation, plasma formation, recoil pressure, and evaporation, can be responsible for the removal of the material.\textsuperscript{12,13} Due to the physical properties of the pulse train and the characteristics of the scanner movement, a multitude of laser and scanner parameters exist, which determine the areal energy input. Nevertheless, it often remains uncertain which of the geometric and energy-related variables describe the process best and how they influence each other as well as the process results.

Statistical methods are a suitable means to examine a large number of parameters simultaneously and systematically. For this reason, they have been used repeatedly in laser materials processing. With regard to ceramics, the response surface method was used by Dhupal \textit{et al.}\textsuperscript{9} and Kibria \textit{et al.}\textsuperscript{10} to optimize the micro-grooving of aluminum titanate and the micro-turning of alumina, respectively. In the following, it is shown how special statistical methods based on approximation models were used to determine the main influencing parameters of the ultrashort pulse ablation of ceramics and to derive general conclusions out of it.

\section*{B. Statistical methods}

To investigate the 2.5D surface ablation of alumina, the software \textit{optiSLang} by Dynardo was used to execute a sensitivity analysis including Design of Experiments (DoE). By definition, a sensitivity analysis serves to determine the correlation between the variance of input variables and the variance of output variables.\textsuperscript{11} The influence of the input variables on the result is identified in order to distinguish between relevant and less relevant parameters. However, existing variance-based methods require huge numerical or experimental effort due to a large number of simulation runs or experiments and are not sufficiently accurate to describe multidimensional problems. Therefore, metamodels are used to compute the responses depending on the input variables with the help of approximation functions. There are several methods to generate metamodels such as polynomial regression, moving least squares or kriging, which minimize the deviations of the predictions compared to the true data points. However, it is often not clear which method is most suitable for which problem.\textsuperscript{12} Therefore, \textit{Dynardo} developed the Metamodel of Optimal Prognosis (MOP), which has been introduced by Most and Will in 2008.\textsuperscript{13} This selects the best combination of input parameters as well as an optimal metamodel with the help of an objective quality measure, the so-called Coefficient of Prognosis (CoP).

The approximation quality of a metamodel is usually estimated with the well-known Coefficient of Determination (CoD, $R^2$). However, this measure behaves too optimistic for a small number of data points or an increasing number of input variables. Furthermore, it is only applicable to polynomials, which makes the use of more complex but possibly more accurate approximation methods difficult. In contrast to that, the CoP is model independent and does not overestimate the approximation quality when the number of samples is relatively small.\textsuperscript{13} It is defined as follows:

$$\text{CoP} = 1 - \frac{SS_P}{SS_T},$$

with $SS_P$ as the sum of squared prediction errors and $SS_T$ as the total variation of the outputs. The errors are estimated by means of cross validation. In this procedure, the data points are divided into a training group for generating the metamodel and a test group for measuring the quality of the model. In this way, the model quality is determined only by the data points which are not used to create the approximation model. As a result of the MOP, an approximation model is obtained that contains the highest CoP for each output parameter as well as the corresponding most important variables.

\section*{II. EXPERIMENTAL APPROACH}

The experiments were carried out with a mode-coupled solid-state laser (Lumera Laser, Hyper Rapid 25) with a pulse duration of 9 ps and a wavelength of 1064 nm. Via external frequency conversion, the second and third harmonic are generated, so that 532 and 355 nm are available as well. The beam, which has a Gaussian shaped intensity profile, is deflected by a galvo scanner and focused on F-Theta objectives of varying focal lengths. To remove the material, test fields of 5 mm $\times$ 5 mm have been filled with parallel scanning lines. The direction of the lines has been randomly rotated after each layer to achieve a homogeneous surface treatment. The evaluation of the ablation process is based on the output parameters profile depth and roughness, which have been measured with the help of a laser scanning microscope (Keyence, VK-X100). The roughness has been determined by a multiple line scan and the profile depth by measuring the step distance between initial and processed surface, as can be seen in Fig. 1. To evaluate the efficiency of the process, the ablation rate was calculated as the ratio of ablated volume and processing time.

Using the so-called Latin Hypercube Sampling (LHS), an experimental design with 100 parameter combinations was set up. This special DoE scheme, which has been described by McKay \textit{et al.},\textsuperscript{14} is especially suitable for the analysis of complex nonlinear systems. The LHS is a

![FIG. 1. Measurement of roughness and ablation depth with achieved value ranges for Al$_2$O$_3$.](image)
stochastic sampling method, in which the parameter settings are spread randomly and uniformly over the whole design space within the lower and upper bounds. By means of stochastic evolution strategies, undesired correlation errors between the input variables are minimized. In contrast to deterministic sampling schemes, such as full factorial or fractional factorial designs, the number of samples does not increase exponentially with increasing dimension and the input values are not limited to only two or three levels in each dimension. Therefore, the LHS can be used to obtain a maximum of information with little effort, even if nonlinear parameter correlations are considered.

Table I gives an overview of the investigated input and output parameters and their value ranges used within the parameter study. While some inputs can be changed directly by the machine settings (actuating variables), others result from physical connections (controlled variables). Within the scope of the sensitivity analysis, all listed parameters have been taken into account to identify their significance for the removal process.

### III. RESULTS AND DISCUSSION

#### A. Sensitivity analysis

The results of the sensitivity analysis based on the MOP can be visualized with the help of the CoP, as shown in Figs. 2 and 3. The bar charts illustrate the significance of the input variables with respect to the responses roughness and ablation depth. The single indices of the input variables refer to their variance contribution and thereby show how much every input affects the particular output. The total CoP, given in the head of the diagrams, quantifies the quality of the full approximation model. Unimportant variables are filtered by the system, so that they do not appear in the CoP chart.

Due to the physical relations between several input parameters, high input correlations occur, which reduce the model quality and make realistic conclusions difficult. For this reason, it has been necessary to filter some parameters manually so that input correlations can be avoided and physically meaningful models of high prognosis ability are generated. In particular, the best models could be created either by considering power, pulse distance, and line distance in combination with the focus diameter (Fig. 2) or fluence and pulse overlap without taking the focus diameter into account, since its influence is already included in the aforementioned parameters (Fig. 3).

It is obvious that fluence and pulse overlap have the highest impact on both output parameters, whereas the wavelength has almost no influence at all. Concerning the ablation depth, the most significant input variable is the number of layers, which was to be expected. Much more interesting is the fact that this parameter has absolutely no influence on the remaining responses, roughness, and ablation rate. This indicates that the ablation process continues constantly into depth, at least within the examined range of 20 layers.
maximum. Each layer can be processed under the same conditions, irrespective of the number of layers which have already been removed. Such a behavior is a prerequisite for the three-dimensional structuring of material with high shape accuracy. As a result, a high agreement of calculated target contours with processed actual contours can be expected.

In addition to the basic knowledge concerning the significance of the inputs related to the outputs, important functional correlations and processing principles can be derived from the sensitivity analysis as well. With regard to the surface quality, it is generally assumed that the roughness increases with increasing fluence or pulse overlap. However, the metamodeling indicates that there exists an optimal pulse overlap, for which the roughness can be reduced. Starting from this point, the roughness increases for both increasing and decreasing overlap. At the same time, the optimum strongly depends on the fluence. This results in several local minima and maxima as can be seen in the response-surface diagram in Fig. 4. In detail, Fig. 5 underlines that the pulse overlap leading to a minimal roughness shift to smaller values if the fluence is increased. This behavior was already observed for the ultrashort pulse ablation of glass ceramics and could now be proven for alumina. It can be explained by the fact that the effective ablation diameter increases with an increase in power. That leads to a larger effective overlap at similar pulse distances. However, since the experiments are based on the theoretical overlap, a shifting of the values occurs. As a result, a small roughness, and therefore a good surface quality is achievable even for high fluences, just by adapting the pulse distance.

The depth-related output parameters ablation depth and ablation rate highly depend on the average laser power. Figure 6 shows that the ablation rate increases steadily with an increase in power. In addition, the pulse distance and the line distance have a decisive influence on the effectiveness of the material removal. The response-surface diagrams reveal that the functional relation between ablation rate and pulse distance significantly changes through a variation of the line distance.

In the case of large line distances [Fig. 6(a)], the removal rate increases with decreasing pulse distance. This is due to the increasing pulse overlap, which generates higher ablation depths. However, the ablation rate is also determined by the processing speed. Since small pulse distances are generated by the use of small scanning speeds, they generally have a negative effect on the removal rate. However, the steady increase in the ablation rate with decreasing pulse distance indicates that this effect plays only a subordinate role.

On the other hand, regarding small line distances [Fig. 6(c)], a completely different functional interrelationship between ablation rate and pulse distance can be observed. Especially regarding high power values, the removal rate now increases with increasing pulse distance, i.e., with decreasing pulse overlap. This seems to contradict the previous observations, but it can be justified by the high influence of the line distance on the ablation rate. It has a considerably higher impact on the process efficiency than the scanning speed. Decreasing line distances, as well as small pulse distances, generate higher pulse overlaps and thus lead to an increase in the removal depth. However, small line distances also mean that the area to be processed has to be filled with more scanning lines. Due to the many jumping and marking vectors to be executed, the processing time increases significantly. For this reason, the ablation rate generally decreases with decreasing line distance. The loss of time at small line distances is so crucial that the scanning speed gains in importance. High marking speeds and resulting high pulse distances can now lead to an increase in the ablation rate.

In between, there exists a line distance for which the opposing influences of the pulse distance on the ablation depth and the processing time compensate each other, so that an independence of the removal rate from the pulse distance can be detected [Fig. 6(b)].

By investigating the ablation process of alumina with the help of a sensitivity analysis, useful new insights into the beam–material interactions of ultrashort pulses with technical ceramics could be gained. However, it is especially
interesting to examine how the material properties affect the ablation process. Therefore, additional approximation models for different ceramic substrates shall be set up within further investigations to find out how the chemical composition and structural differences, e.g., grain size and density, affect the surface quality and the efficiency of the material removal. An initial step according to that has been already made by investigating the ablation depth depending on the pulse overlap for various ceramic materials with all other process parameters held constant. For this experiment, for which the results are given in Fig. 7, the horizontal and the vertical pulse overlaps have been equated. The graphs show that all materials follow the same nonlinear behavior, whereas the exact values differ a lot. The industrially important aluminum nitride ceramic (AlN) exhibits the smallest material removal of all examined substrates. Alumina, porcelain, and the investigated low-temperature co-fired ceramic (LTCC), which is a glass ceramic mainly consisting of borosilicate glass and alumina, show almost the same ablation behavior. It is assumed that this can be attributed to similar structural properties. In consistency to that, by far the highest material removal can be achieved if the ceramics are in the green state, as can be seen in the case of porcelain and LTCC.

The similar behavior of the different ceramics regarding the functional correlations between the process parameters indicates that the knowledge collected within the sensitivity analysis can be applied to a wide range of materials. Based on this, the achieved results were used to deal with industrially interesting applications and to optimize them in terms of quality and efficiency.

B. Applications

In industry, technical ceramics are used for various products. In the following, two fundamentally different applications are presented to exemplify, which benefits the ultrashort pulse technology has for the processing of ceramics. It is shown how the disadvantages of conventional laser processes can be overcome, without losing efficiency.

1. Separation of Al₂O₃ substrates

Especially in the semiconductor industry, ceramics are applied as a base material for a huge amount of devices. Due to the increasing demand of assembly-free electronic devices, fast and accurate cutting techniques for technical ceramic substrates are in great demand. A fast cutting method is mechanical dicing, which often results in mechanical fracture and large kerf widths. Another common way uses CO₂ and Nd:YAG lasers, mainly in combination with scribing processes.

Figure 8(b) exhibits the resulting problems with a CO₂ laser scribing process. The longitudinal section reveals long cracks and debris on the top. Due to the long pulse and the high energy input, the laser affected zone also shows glazing. In comparison to that, ultrashort pulse lasers can minimize the HAZ and the collateral damage of the surrounding material. Concerning semiconductor usage, the cleaning and burring process is very important. The aim of the investigations was to achieve a significant improvement, reducing the steps and the processing time in general.

The optimized process strategy [Fig. 8(a)] shows no glazing, cracks, and debris. For this reason, the final cleaning steps can be eliminated. An additional optimization of the...
process is the material usage. Picosecond laser pulses can machine with minimal groove width. Regarding a standard chip, the material usage could be increased from 64% to 90% by using the picosecond laser grooving instead of the CO2 laser scribing. For a material thickness up to 250 $\mu$m, the ultra-short pulse processing exhibits the same processing time and the same fracture force. For higher thicknesses starting from 500 $\mu$m, the laser process is slower, but the whole processing time including cleaning steps is still more efficient.

2. Precision structuring of porcelain surfaces

In addition to their usage in high-tech industries such as the semiconductor and electronics industry, ultrashort pulse lasers can be also applicable for innovations in consumer goods. Thus, investigations to structure precise 2.5D designs into different porcelain surfaces of high-quality crockery have been conducted. In addition to design aspects, such structures can serve to individualize or counterfeit-proof such products.

TABLE II. Properties of ultrashort pulse laser ablated porcelain surfaces.

<table>
<thead>
<tr>
<th>Surface-type</th>
<th>Initial roughness</th>
<th>Processed roughness</th>
<th>Transition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Biscuit</td>
<td>$Ra = 2 \mu$m</td>
<td>$Ra = 1.5 \mu$m</td>
<td></td>
</tr>
<tr>
<td>Glazed</td>
<td>$Ra &lt; 0.1 \mu$m</td>
<td>$Ra = 1.5 \mu$m</td>
<td></td>
</tr>
<tr>
<td>Green</td>
<td>$Ra = 2 \mu$m</td>
<td>$Ra = 4.4 \mu$m</td>
<td></td>
</tr>
<tr>
<td>Fired</td>
<td>$Ra = 2.2 \mu$m</td>
<td>$Ra = 4.2 \mu$m</td>
<td></td>
</tr>
</tbody>
</table>

With the exception of the possibility to glaze porcelain surfaces with the aid of CO2 lasers, it has not been possible to bring clean, deep profiles into porcelain neither with CO2 nor with Nd:YAG lasers so far. The processing of porcelain with solid-state lasers of long pulse durations leads to brittle edges of high roughness and an ablated surface that is strongly affected by material deposits. In addition to the highly precise material removal that can be achieved with the aid of ultrashort pulse lasers, a further advantage of this method is that it is nearly independent of the initial surface texture. Table II shows how various porcelain materials have been ablated by means of picosecond pulses. For all of them, the biscuit porcelain, the glazed substrate, and the raw material, a defined, crack-free transition from untreated to processed surface can be observed. Comparing glazed and unglazed porcelain, a similar roughness is generated, which results in identical visual and haptic properties. Only the ablation of the raw material leads to a significantly higher roughness considering the same set of parameters. However, it is particularly interesting that the laser processing does not adversely affect the properties of the unfired material. When the structured substrate is fired, no cracks or geometrical changes occur. The roughness produced in the green state is retained during the firing process.

The various possibilities of structuring offer complete freedom to the product designer. The surface profiles can be applied to different material conditions before or after the firing process. Moreover, the structuring of metallizations such as gold coatings is possible without material damages as well. The resulting process qualities are shown in Fig. 9, using the example of a filigree logo.

IV. CONCLUSION

Within the scope of the investigations, it could be shown that a sensitivity analysis based on metamodeling can be used to identify the most significant parameters as well as physical relations between input and output variables of complex laser processes. The gained knowledge can be used to optimize efficiency and quality of the process and to develop general processing strategies. It should be noted, however, that the models must always be thoroughly examined regarding their physical validity. In particular, a filtering of input parameters has to be executed in order to avoid input correlations and to obtain well-founded models.

Fluence and pulse overlap have been identified to be by far the most important influencing parameters, which describe the process best. The results concerning the surface quality...
show that it is possible to achieve acceptable roughness values even at high laser powers by optimizing the pulse overlap. Regarding the efficiency of the process, it makes sense to consider pulse distance and line distance separately in order to generate the highest possible ablation rates. This contradicts the so far existing strategy of always equating pulse distance and line distance in favor of a homogeneous energy distribution.

The shown applications illustrate that ultrashort pulse laser processing can enable a significant increase in quality compared to conventional processing methods in the ceramics industry. The processing time can be minimized by process optimization so that it is not inferior to that of other laser processes. Nevertheless, further detailed investigations have to follow. By including other technical ceramics into the metamodeling, the influence of the material shall be examined in order to gain an even better understanding of ultrashort pulse laser processes.

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