

Available online at www.sciencedirect.com





Physics Procedia 5 (2010) 311-316

www.elsevier.com/locate/procedia

Investigation on particle formation during laser ablation process

LANE 2010

T. Scholz^a*, K. Dickmann^a

with high brilliant radiation

^aLaser Center (LFM), Münster University of Applied Sciences, Steinfurt, Germany

Abstract

Nanometer sized particles are formed within the vapor plume during the ablation of metal with laser radiation. Thereby, the particle formation rate depends strongly on the used intensity of the laser source. High brilliant laser sources have the ability to generate intensities higher than $1\cdot10^8$ W/cm² during cw operation. Due to the widespread use of high brilliant laser sources in research and industrial applications, it is important to investigate the influence of particle formation on the ablation process. Therefore, the presented work is focused on the particle formation during the ablation process of stainless steel with a single-mode fiber laser. Results of experimental work are shown and analyzed. In the experimental work a probe laser beam is directed through the ablation plume and the scattered intensity is analyzed. TEM images of particles show an average particle size of 9 nm at an intensity of $1.92 \cdot 10^8$ W/cm².

© 2010 Published by Elsevier B.V. Open access under CC BY-NC-ND license.

Keywords: ablation; particle formation; brilliant laser sources; cw operation; scattering

1. Introduction

The interaction between laser radiation and resulting vapor plume impacts laser material processing. Matsunawa describes that scattering and absorption of radiation by ultra-fine particles influence the intensity on the material surface [1]. Thereby, the particles are formed due to condensation within the vapor plume. After starting the process radiation is absorbed at the material surface. Melt is formed and a part of the ablated material vaporizes. The vaporized material propagates as a shockwave. At this moment the system is in a supersaturated state and phase change within the vapor is induced by particle formation. Nucleation starts and particles are formed due to collision in the vapor plume. Fundamental work on particle formation and processes is presented by Smirnov [2].

Various authors describe the particle formation within the expanding vapor plume during laser ablation [3-8]. Kar et al. present a numerical model to analyze the particle formation during the ablation process of niobium [3]. The

^{*} Corresponding author. Tel.: +49-2551-962328; fax: +49-2551-962490. *E-mail address*: scholzt@fh-muenster.de.

regarded process is based on the ablation with a KrF excimer laser. The wavelength is 248 nm, the pulse duration 30 ns and the intensity is varied between 499 and 833 MW/cm². It is significant that with increasing duration the mean particle size rises up. Also, the particle size increases with longer distance from the surface. Gnedovets et al. concentrate on the ablation process with lower intensities [4]. They relate to a Nd:YAG laser with a pulse duration of 1 ms and an intensity of $4 \cdot 10^4$ W/cm². The results show that there exists a thin layer near the material surface where no particles are formed. The reason is due to the fact that the vaporized material condenses on the surface of the material. By regarding on the particle size distribution in relation to the distance from the material surface it can be seen that the mean particle size decreases with the distance from the material. The influence of ionization on particle formation is analyzed by Tillack et al. [5]. The basis of the presented investigation is the ablation process of silicon with a Nd:YAG laser featuring a pulse duration of 8 ns and a wavelength of 532 nm. The intensity is varied between $5 \cdot 10^7$ and $5 \cdot 10^9$ W/cm². A direct correlation between intensity, particle density and degree of ionization is shown by the results.

Goncharov et al. present an experimental setup to investigate scattering behavior, absorption and transmission of the vapor plume [9, 10]. They relate to the ablation process of zinc with a Nd:YAG laser. The intensity is $1.2 \cdot 10^6$ W/cm² and the pulse duration 700 μ s. Furthermore, the beam of a ruby laser is directed through the vapor plume. The entire experimental setup is located within an integrating sphere and the transmitted and scattered signals are detected by a photodiode. Finally, the absorption, scattering and transmission are calculated by using the Rayleigh theory.

Compared to previous research, the presented work is focused on the particle formation during the ablation process of stainless steel under cw operation. Two intensities are used: $0.64 \cdot 10^8$ W/cm² and $1.92 \cdot 10^8$ W/cm². Thus, the intensities are in a comparable parameter range as the intensities of short-pulse lasers. But in contrast to the ablation with short-pulse lasers, the particle formation takes place during the irradiation and influences the ablation process.

2. Experiment

2.1 Fiber laser source

A single-mode fiber laser was used for the experimental setup. Table 1 presents the characteristic data of the laser source.

Туре	IPG, YLR-300SM
laser power	≤300 Watt
focussed beam diameter	14 µm
wavelength	1070 nm
M^2	≤1.1
pulse rep. rate	< 50 kHz

Table 1. Beam and pulse parameters of applied fibre laser source

The beam is focused on the material surface with a maximum intensity of $1.92 \cdot 10^8$ W/cm². Within the experimental work, the ablation duration is 100 μ s.

2.2 Particle size distribution

For experimental measuring of the particle size distribution, a metal sheet is positioned between two glass plates, figure 1. The beam is focused on the edge of the metal sheet and the particles are deposited onto the surface of the

glass plates. Copper grids with Formvar or Carbon coatings are added in the experimental setup. They are anchored on the glass plates and investigated with a transmission electron microscope (TEM).



Fig. 1. Experimental setup for measuring the particle size distribution during the ablation process

As part of further investigations, the beam of a probe laser is directed through the vapor plume. It orthogonally intersects the optical axis of the fiber laser beam. Due to particle formation within the vapor plume, the radiation of the probe laser is scattered. The scattered intensity depends on the particle size and density distribution in the vapor plume. The probe laser is a HeNe laser with a wavelength of 543 nm and an output power of 1 mW. A Si photodiode is used to detect the scattering intensity. It is fixed on a rotating table to enable an angular alignment between 0° and 90°. Figure 2 illustrates a schematic of the experimental setup.



Fig. 2. Schematic of the experimental setup for measuring the angle-dependent scattering of the radiation of a HeNe laser. The beam of the fiber laser impacts vertically on the metal sheet. Within the interaction zone the beam of the HeNe laser crosses the optical axis of the beam of the fiber laser in an orthogonal way

2.3 Ablation velocity

For verification of the influence of interaction between clusters and laser radiation on the ablation velocity, the ablation velocity is indirectly measured. Therefore, the drilling duration through metal foils with a known thickness are measured as the delay between two photodiode signals. One photodiode is positions above the material and detects the start signal. The other photodiode detects the signal under the treated metal foils. The ablation velocity results from the relationship between the material thickness and the delay between the two photodiode signals.

3. Results and discussion

Figure 3 presents a TEM image of particle structures which were deposited on the surface of the copper grid. The formation of these structures cannot be attributed to melt ejection. It is pointed out that the fine structures consist of spherical particles. The particles were formed during the ablation process. Subsequently, the condensation stops and the formed particles agglomerate after collision due to van der Waals and electrostatic forces. Wen et al. describe a similar formation [8]. They observed the agglomeration of particles after the ablation process of copper with a short pulsed Nd:YAG-Laser. Figure 4 presents a TEM image of particle structures which are ascribed to the ablation process with an intensity of $0.64 \cdot 10^8$ W/cm².



Fig. 3. TEM image of particles. They were formed during the ablation process of stainless steel with a single mode fiber laser (intensity of 1.92·10⁸ W/cm²)



Fig. 4. TEM image of particles (intensity of 0.64·10⁸ W/cm²)

Here, particles are also displayed. But the agglomeration is not as pronounced. It can therefore be concluded that the particle density is not as high as during the ablation with an intensity of $1.92 \cdot 10^8$ W/cm². Figure 5 shows the experimentally determined particle size distributions. They were obtained by evaluation of TEM images. Comparing both distributions, higher particle sizes follow from the ablation with an intensity of $1.92 \cdot 10^8$ W/cm²: A maximum

314

particle size of 29 nm is reached, whereas at an intensity of $0.64 \cdot 10^8$ W/cm², only particle sizes up to 11 nm are measured. Even the width of the distribution is not as wide as at the higher intensity.



Fig. 5. Experimentally determined particle size distributions for two intensities. The distributions are given as relative frequency

Figure 6 presents the ablation velocity and the scattered intensity vs. ablation depth for the intensity of $1.92 \cdot 10^8$ W/cm². The scattered intensity was detected by a Si photodiode.



Fig. 6. Ablation velocity and scattered intensity of the radiation of a HeNe laser vs. ablation depth. The radiation of the HeNe laser is scattered due to particles within the vapor plume. The angle refers to the optical axis of the beam of the HeNe laser. The scattered intensity was detected by a Si photodiode

It points out that the ablation velocity and scattering intensity show opposite trends. The scattering intensity decreases with increasing ablation depth, whereas the ablation velocity increases until 40 μ m. After that, the ablation velocity decreases and goes up to a maximum of 33 m/s at an ablation depth of 100 μ m. According to the Rayleigh theory [11], this result infers a higher particle density and average particle size within the interaction zone at the beginning of the ablation process.

4. Conclusion and outlook

Within the presented work, the particle development during the ablation process of stainless steel with a singlemode fiber laser is investigated. Experimental researches are carried out. A probe laser beam is directed through the vapor plume and the scattered radiation is analyzed. Furthermore, glass plates and copper grids are integrated within the experimental setup. After the process, particle formations are analyzed by TEM images to determine the particle size distributions.

The analysis of the experimental work offers that the particle densities and sizes reach higher values at higher intensities of the fiber laser. Concerning to the ablation with an intensity of $1.92 \cdot 10^8$ W/cm², the experimental results show a maximum of the particle size distribution at 9 nm, a FWHM of 5 nm and a maximum particle size of 29 nm. At the intensity of $0.64 \cdot 10^8$ W/cm², the experimental results present a maximum of the particle size distribution at 6 nm, a FWHM of 3 nm and a maximum particle size of 11 nm. The comparison between the course of the ablation velocity and the scattering intensity shows a strong influence of cluster formation at the beginning of the ablation process.

Relating to applications in industry and research, the results of the presented work show an increase in particle formation during the ablation with high brilliant radiation. The reason lies in the combination between the used material, high intensity and quasi-cw operation. Consequently, in some cases, interaction effects could influence the ablation process and thus the ablation velocity or processing quality.

Acknowledgements

The authors gratefully acknowledge the financial support of the German research foundation (DFG) within grant DI 456/10-1.

References

[1] A. Matsunawa 1990 Physical Phenomena and their interpretation in Laser Materials Processing. In: Int. Conf. on Laser Materials Processing ICALEO, pp 313-324.

[2] B. M. Smirnov 2010 Cluster processes in gases and plasmas Weinheim: WILEY-VCH.

[3] Kar A and Mazumder J 1994 Mathematical model for laser ablation to generate nanoscale and submicrometer-size particles *Phys. Rev. E* 49 410-419.

[4] A. G. Gnedovets, A. V. Gusarov, I. Smurov 1999 A model for nanoparticles synthesis by pulsed laser evaporation J. Phys. D: Appl. Phys., 32, 2162-2168.

[5] Tillack M S, Blair D and Harilal S 2004 The effect of ionization on cluster formation in laser ablation plumes Nanotechnology 15 390-403.

[6] Perrière J, Boulmer-Leborgne C, Benzerga R and Tricot S 2007 Nanoparticle formation by femtosecond laser ablation *J. Phys. D: Appl. Phys.* **40** 7069-7076.

[7] Lescoute E, Hallo L, Hébert D, Chimier B, Etchessahar B, Tikhonchuk V T, Chevalier L M and Combis P 2008 Experimental observation and modeling of nanoparticles formation in laser produced expanding plasma *Phys. Plasmas* 15 063507.

[8] Wen S B, Mao X, Greif R and Russo R E 2007 Experimental and theoretical studies of particle generation after laser ablation of copper with a background gas at atmospheric pressure J. Appl. Phys. 101 123105.

[9] Goncharov V K 2001 Action of high-energy Neodymium laser radiation pulses having a different space-time shape of metals J. Eng. Phys. Therm. 74 1173-1187.

[10] Goncharov V K, Min'ko L Y and Nasonov V I 1983 Optical properties of laser-erosion plasma torches J. Appl. Spect. 39, 1381-1384.
[11] Miles R B, Lempert W R and Forkey J N 2001 Laser Rayleigh scattering Meas. Sci. Technol. 12 R33-R51

316