Evaluation of Crater Width in Nanosecond Laser Ablation of Ti in Liquids and the Effect of Light Absorption by Ablated Nano-Particles

Marzieh Akbari Jafarabadi and Mohammad Hossein Mahdieh[*]

Department of Physics, Iran University of Science and Technology, Tehran, Iran

[*]**Corresponding Author Email:** mahdm@iust.ac.ir

ABSTRACT— **Micro size craters were created by interaction of nanosecond laser beam with titanium target in liquid media. The dimension of crater i.e. depth and width is important in some applications such as micromachining. When the interaction occurs in liquid environment, the ablated materials from the target expand into the liquid. The ablated material can affect the interaction process if the ablated material concentration increases. In this paper, we study the effect of ablated materials in liquids on the crater width. The crater dimension was characterized by using an optical microscope. The results show that not only the type of environment liquid is important in the final size of the created craters, but also the laser fluence and the liquid depth in which the interaction takes place is important in the crater size.**

KEYWORDS: Crater width, High power pulsed laser, Laser ablation, Liquid ambient, Titanium target, Ablated material.

I. INTRODUCTION

Laser induced ablation has been used in the last decades as a capable method in industry and medicine because of its extending unique advantages. Micromachining, nanoparticle production, pulsed laser deposition and material identification are some examples of the most widely used applications of laser ablation in the industry [1]–[4].

Laser ablation is usually induced by focusing a high power pulsed laser beam on the surface of a target material. A portion of the beam energy is absorbed by free electrons on the target surface and the material heats up as a consequence of electron-phonon interaction. In low fluences, the target surface is only melted and evaporated. The melted material flows due to the temperature gradient on the surface and as a result, the surface structure modifies. It has been found that under different conditions of laser pulse duration, different kinds of micro and nano structures can be formed at the irradiation ragion [5], [6].

If the laser fluence is sufficiently high, a plasma plume can be formed and the material is removed from the surface as excited electrons, ions and neutrals. The hot plume expands and (except in vacuum interaction medium) generates a shock wave that expands with supersonic velocity toward the laser beam [7]–[9]. Therefore, as a consequence of considerable material removal, a crater forms at the irradiation region. Several processes such as normal boiling, homogeneous boiling and bubble formation may occur depends on laser beam characteristics, target material properties and interaction ambient [10]–[14].

Nanoparticle production and micromaching by laser ablation in liquid as an interaction medium have been interested in recent years because of some advantages of liquid with respect to air [15]–[23]. Synthesizing of nanoparticles by using laser ablation in a liquid medium is much simpler and easier procedure compared to the conventional chemical methods [15], [16]. The possibility of controlling the size of the produced nanoparticles is also a prominent feature of this method. Material processing with laser ablation can be strongly influenced by laser beam parameters such as wavelength, energy, beam spot width, number of pulses and pulse duration [11]–[13]. The properties of ambient medium are also very effective at the crater morphology, depth and width [14], [17]–[23]. Due to higher pressure and temperature of the plasma in ambient liquid with respect to those in ambient air or vacuum, craters with greater depths and widths can be formed in laser ablation in ambient liquid. The liquid environment can also result in the elimination of debris redeposition at the irradiated region.

Many researchers devoted their research to study laser ablation of different solid targets in liquid medium. For example, by ablating silicon target with a KrF excimer laser, Zhu *et al.* obtained higher values of crater depth for ambient water in compare with those in ambient air [18]. In our previous work, we also investigated the effect of pulse number and laser intensity in laser ablation of aluminum targets in both air and water [23].

It must be noted that the control of crater width is an important issue in material processing with laser ablation. In the case of laser ablation in liquid ambient, the ablated material expands into the liquid medium during the irradiation time and thereafter. Practically, irradiating the target with the first few pulses leads to a small increase in the concentration of the ablated (removed) material in the liquid medium. However, if the interaction takes place with higher number of pulses, the amount (concentration) of the removed material in the liquid increases. Therefore, in the condition of irradiation with high pulse numbers, the laser beam may reach the target surface after passing through liquid with highly concentrated micro/nano size ablated particles. In such condition part of the laser beam energy gets absorbed by these particles before reaching the target surface. Obviously, the absorbed energy is related to the concentration of the suspended micro/nano particles in liquid. As a result, some issues such as the heat affected zone (HAZ), and the geometrical features of the crater on the target surface may be significantly influenced by the ablated micro/nano particles.

To our best knowledge, there is no report to show the effect of absorption of light by the ablated material on crater dimensions i.e. crater depth and width.

In this paper, we studied experimentally the influence of the amount of the ablated material on the crater widths. The ablation process occurs during the irradiating of a titanium target by Nd:YAG high power pulsed laser in three different environment i.e. distilled water, acetone and ethanol. In a separate experiment the irradiations took place at a fixed laser fluence while the target was placed at different depths in distilled water. The experiment was also performed for different laser fluences in distilled water and a constant water depth. The results show that the crater width is very sensitive to the amount of energy received on the target surface.

II. EXPERIMENTS

Figure 1 shows a schematic diagram of the experimental setup. A high power pulsed Nd:YAG laser with 1064 nm wavelength, 10 ns pulse duration and 10 Hz repetition rate was used to induce ablation. The Nd:YAG laser beam was initially expanded by a beam expander and focused on the target surface by a doublet lens (*f* ~ 18 cm,) providing a beam spot radius of ~ 150 µm at the target surface. As this figure shows, in each irradiation a small portion of the beam was reflected by a splitter and the beam energy was recorded by a pyroelectric energy meter. Titanium with the thickness of 3 mm was irradiated in three liquid media (distilled water, acetone and ethanol). In order to perform experiments in liquid medium, the titanium target was placed in a small cell.

In each irradiation, the titanium target was exposed to the laser beam within a certain time in which the total number of pulses irradiated on the target surface was 1500. In the present paper, the effect of ablated material concentration (in liquid) on crater width was studied while different laser fluences and liquids were used in the experiments. In the experiment, firstly the titanium target was irradiated with specific laser fluence (and with

1500 shots) while the target was placed in a specific depth (of the ambient liquid). Then the target moved to a fresh region and the irradiation was repeated again. In the second irradiation one can assume that the ablated material (of the first irradiation) absorbs part of the incident laser beam energy, and reduce the loaded energy at the target surface. This process was repeated for eight fresh regions of the target surface. It must be noted that at each step, the accumulation of removed material in the liquid medium increases. Therefore the transparency of the liquid medium reduces more after each step.

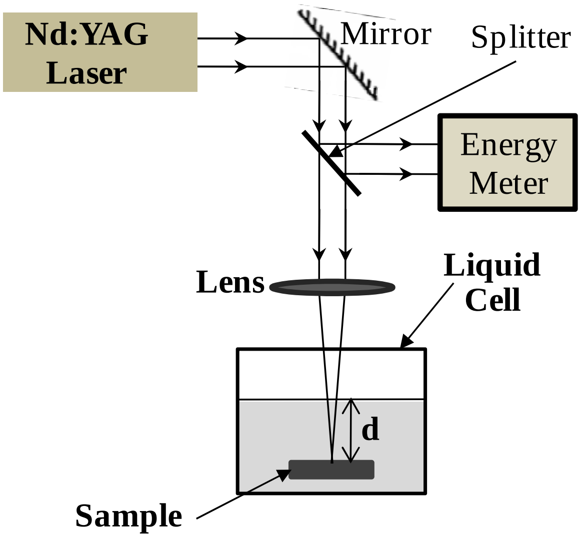

Fig. **1** Schematic diagram of the experimental setup.

For estimating the amount of ablated material, a cone shape geometry was assumed for the crater. There are some reports indicating the creation of craters with almost cone geometry by nanosecond laser ablation of metals and semiconductors [23], [24]. Therefore, the crater volume can be calculated based on the crater dimensions (depth and width). In our experiment, the depth of produced craters was measured by using an optical microscope with maximum magnification of 1000×. The microscopic images of the craters were analyzed by image processing software and the craters' width were characterized. The amount of the removed material from each crater can be estimated when the crater volume is known.

In the experiments, we focused our study to evaluate three different issues which affect the crater characteristics. First, we studied the effect of laser fluence delivered on the target surface (without considering the light absorption by ablated materials and environment liquid) at a constant water depth (the distance "d" in Fig. 1 in which the titanium target is placed). In the second attempt the irradiation was performed in distilled water at a constant laser fluence (without considering the light absorption by ablated materials and environment liquid) and the effect of water depth was investigated. In the third experiment, three liquid material (distilled water, acetone and ethanol) were examined and the laser fluence (without considering the light absorption by ablated materials and environment liquid) and liquid depth were kept constant (55 J/cm^2 and 10 mm respectively).

III. RESULTS AND DISCUSSION

Typical images of the craters on the titanium surface are shown in Fig. 2. The data for this figure are summarized in table 1. Each crater was produced by irradiation of the titanium target surface (total pulses of 1500). The results show that the crater width is varied for different conditions of laser fluence and liquid ambient.

Table **1** The information about the target surface images in Fig. 1.

Figure	Fluence (J/cm^2)	Liquid depth (mm)	Width (µm)	Liquid material
2(a)	90	10	338	water
2(b)	110	7.6	364	water
2(c)	55	10	303	water
2(d)	55	7.6	547	water
2(e)	55	10	265	ethanol
2(f)	55	10	282	acetone

According to the results shown in Fig. 2 and table 1, the crater width is strongly depends on the laser beam characteristics and interaction ambient. There are great differences between the results when the laser fluence or the ambient parameters change. More detailed analysis of the results is discussed in the following sections.

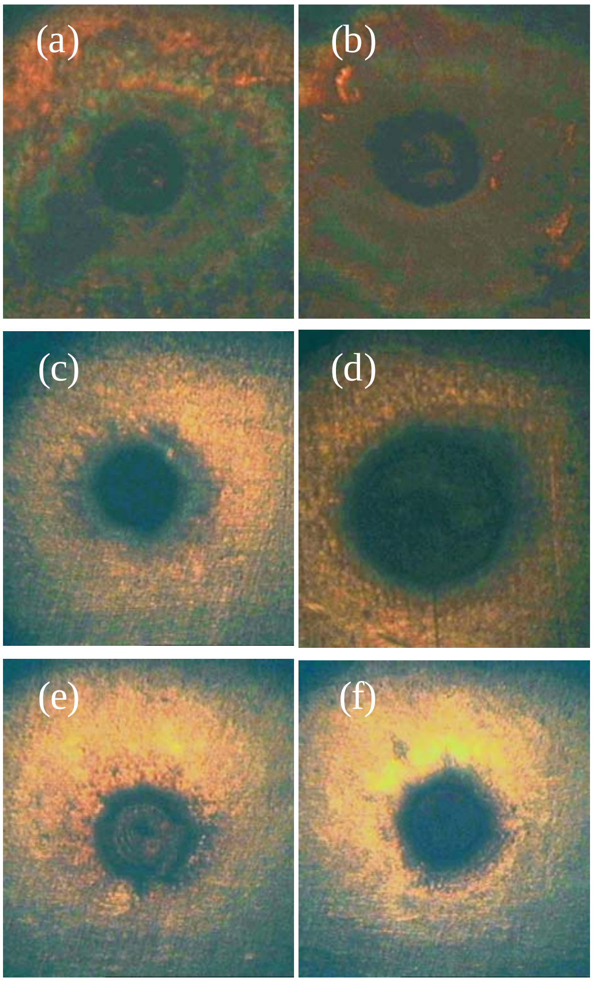

Fig. **2** Typical microscopic images of the craters formed on the titanium surface. The data of the figure are summarized in table 1.

A. *The Effect of Laser Fluence*

In order to investigate the effect of laser fluence on the crater width, the experiment was performed using different laser fluences but keeping the target surface at a constant depth in liquid distilled water (d=10 mm in Fig. 1). Figure 3 shows the crater width versus the relative amount of ablated material at four different fluences 55, 90, 110 and 130 J/cm^2. The results in Fig. 3 show that the crater width increases with laser fluence. For instance, from Fig. 3 it can be seen that the minimum crater width of 470, 410, 390 and 320 μm were obtained for the laser fluences of 130, 110, 90 and 55 J/cm^2 respectively. These results can be explained by the following description. At a constant water depth, the energy loaded on the target surface is increased with increasing the incident laser fluence. Practically, the total spatial part of the beam can not contribute in laser ablation. The ablation can take place if the laser intensity is greater than a threshold intensity. In fact, in each irradiation only part of the beam (where the intensity is enough for ablation) can ablate the target surface. By increasing laser fluence, the laser intensity at a larger area become enough to ablate the material, therefore larger crater can be created. As a result, higher laser fluence leads to the creation of craters with higher widths.

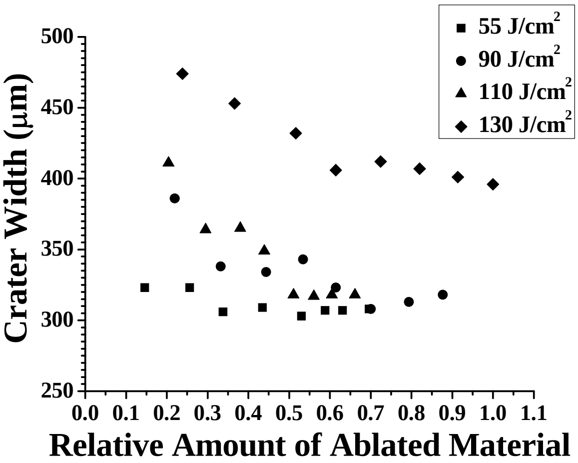

Fig. **3** Crater width versus relative amount of ablated material for 55, 90, 110 and 130 J/cm^2 laser fluences and 10 mm water depth.

The crater width also decreases with increasing of the relative amount of ablated material for any specific laser fluence. As the Fig. 3 shows, the craters width decreases significantly for lower amounts of ablated material and remains almost constant for higher amounts of removed material. The light absorption relates to the properties of the ambient in which the light travels through. Increasing absorbing species in the liquid medium, results in reduction of transparency of the liquid medium. During the ablation process, the plasma plume expands and ablated material (including liquid droplets,) micro and nano particles are ejected from the surface with high velocity [25]. The expanding hot material cools down later and the ablated material spreads out in the liquid medium. In this case, a laser beam must travel through the liquid which is enriched by ablated material. As a result the beam energy is absorbed and dispersed by the ablated particles. Thus, the energy delivered on the titanium surface is reduced. In our experiment, when the first crater is produced, the concentration of the ablated micro/nano particles in the liquid is low, therefore, their absorbing effect can be ignored. However, for the next craters, the laser beam propagates through a more opaque liquid. As a result, part of the beam energy gets absorbed and lower energy is loaded on the target surface. Therefore, a crater with smaller width is formed. It can be concluded that the crater width decreases with increasing the number of craters that are created. However, the absorbing rate decreases with exposure time. This can be seen in Fig. 3 which shows the crater widths remain approximately constant at higher amounts of ablated material.

B. *The Effect of Liquid Depth*

Figure 4 shows the crater width versus the relative amount of ablated material at different liquid depths. Figure 4 shows the results of an experiment in which a titanium target was irradiated in distilled water at a constant laser fluence 55 J/cm^2. The water depth was also changed within a range of 6.7, 7.6, 8.5, and 10 mm to investigate the effect of water depth on crater width.

According to the results, the crater width increases dramatically with decreasing the water depth from 10 to 7.6 mm. Such results may be due to the consequence of decreasing of the laser transmission distance in water. However, the crater width decreases slightly with decreasing the water depth from 7.6 to 6.7 mm. A small reduction in crater width by increasing the amount of removed material is also seen in each water depth. The interaction medium (distilled water) has a high absorption coefficient at the wavelength of laser (~ 0.5 cm^{-1} at 1064 nm). As a result, the energy received by the target surface was restricted to lower value when the water depth is increased. Therefore, the crater width reduces when the target is placed in deeper water. The other reason for such results may be explained by taking into account the absorption of the ablated particles in the passage of the beam. By increasing the water depth, the distance in which the absorbing species absorbs the beam energy increases. Such condition may lead to decrease the received energy on the target surface.

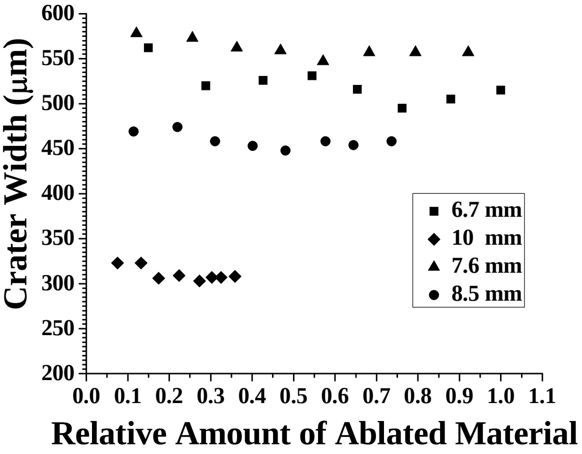

Fig. **4** Crater width versus relative amount of ablated material for 6.7, 7.6, 8.5 and 10 mm water depths at 55 J/cm^2 laser fluence.

On the other hand, in lower water depths the expansion of the plasma and the ablated material are limited to a smaller distance rather than that in a higher water depth. Therefore, plasma with higher temperature and electron density is formed at a lower water depth. Since the trailing edge of the laser beam can be absorbed more considerably in hot and dense plasma than cold and dilute plasma, higher values of energy reached on the target surface at the higher water depth.

As shown in Fig. 4, crater width decreases when the water depth is decreased from 7.6 to 6.7 mm. The data for 6.7 mm depth cannot be explained easily. The results for this depth could be very similar to that of 7.6 mm depth and such similarities could be due to the saturation of the beam fluence at depth higher than ~ 7 mm. As explained earlier, three factors are effective in the energy portion that reaches on the target surface: the distilled water absorption (at the laser wavelength), the absorption of the beam energy by ablated particles (and material) and finally plasma absorption of the laser beam (the trailing edge). The first two issues result in decreasing the crater width at higher water depths, while the last factor is more effective at lower water depths. According to these results, in the condition of our experiment if craters with larger widths are preferred, 7.6 mm water depth is the best option.

C. *The Effect of Liquid Material*

Crater widths with respect to the relative amount of ablated material in water, acetone and ethanol (as the interaction ambient) are shown in Fig. 5. In this experiment the laser fluence and liquid depth were 55 J/cm^2 and 10 mm, respectively.

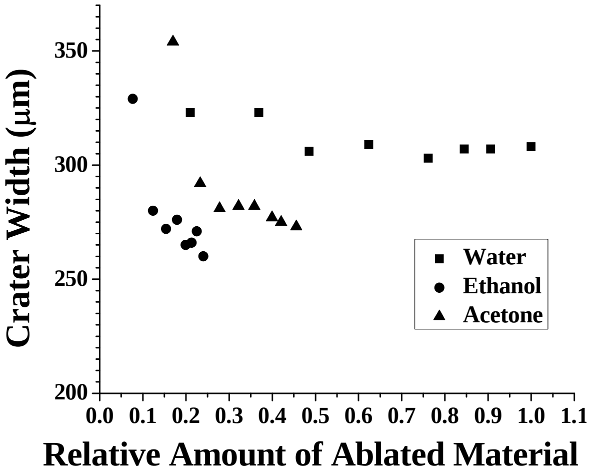

Fig. **5** Crater width versus relative amount of ablated material for three different liquid medium (water, acetone and ethanol) as interaction medium at 55 J/cm^2 laser fluence and 10 mm liquid depth.

Our results show that the crater widths have almost similar trends for three liquid media, i.e. fast reduction in the condition of small ablation and slow reduction (remaining almost constant) for the case of large ablation. However, the amount of material that removed from the titanium surface in water is almost two and five times higher rather than those for acetone and ethanol, respectively. Such results show that more efficient ablation can be achieved in water. Enhancement of the laser ablation in water with respect to those in two other liquids may be due to the consequence of different ablation dynamic processes. The dynamic processes include plasma formation and expansion, ablated material expansion, and shock wave development in liquids.

Excluding the data for the lowest amount of the ablated material, it can be seen from Fig. 5 that the crater widths have larger values in water (as an interaction environment) with respect to those in acetone and ethanol. Such results may be due to the consequence of different absorption coefficients in these liquids at the laser wavelength. Further investigations show that the size (and size distribution) of produced micro/nano particles is different when different liquids are used. Such different sizes of produced particles may be considered as the second reason of such behavior.

IV. CONCLUSION

Controlling the crater width in laser ablation is very important in some industrial applications such as micromachining. In this paper, we investigated the effect of the ablated material (in liquids) on the crater width. The effects of laser fluence as well as interaction medium on the crater width were evaluated. Titanium target was placed in a liquid cell and the irradiation was provided by focusing an Nd:YAG pulsed laser beam on the target surface. From the results it can be concluded that the concentration of the ablated material in the liquid medium strongly affects the light absorption during its propagation through the liquid ambient. From the results we also concluded that greater values of crater width were obtained at higher laser fluences. It was also concluded that in our experiment condition, a distance of 7.6 mm was the optimum water depth to reach the maximum width. The results of this experiment show that the type of the liquid medium also effective on the crater width.

REFERENCES

[1] G. Giusti, J. Bowen, Q. Ramasse, G. Rey, E. Blackburn, L. Tian, I.P. Jones, and J.S. Abell, "Dielectric properties of pulsed-laser deposited indium tin oxide thin films," Thin Solid Films, Vol. 524, pp. 249–256, 2012.

[2] M. Dong, J. Lu, S. Yao, Z. Zhong, J. Li, and W. Lu, "Experimental study on the characteristics of molecular emission spectroscopy for the analysis of solid materials containing C and N," Opt. Express, Vol. 19, pp. 17021–17029, 2011.

[3] B. Kumar, and R.K. Thareja, “Synthesis of nanoparticles in laser ablation of aluminum in liquid,” J. Appl. Phys., Vol. 108, pp. 064906 (1–6), 2010.

[4] M.R.H. Knowles, G. Rutterford, D. Karnakis, and A. Ferguson, “Micro-machining of metals, ceramics and polymers using nanosecond lasers,” Int. J. Adv. Manuf. Technol. Vol. 33, pp. 95–102, 2007.

[5] D. Vipparty, B. Tan, and K. Venkatakrishnan, “Nanostructures synthesis by femtosecond laser ablation of glasses,” J. Appl. Phys. Vol. 112, pp. 073109 (1–7), 2012.

[6] M. Sobhani, and M.H. Mahdieh, “Comparison of sub-micro/nano structure formation on polished silicon surface irradiated by nanosecond laser beam in ambient air and distilled water,” Laser Part. Beams, Vol. 31, pp. 465–473, 2013.

[7] S.S. Harilal, G.V. Miloshevsky, P.K. Diwakar, N.L. LaHaye, and A. Hassanein, “Experimental and computational study of complex shockwave dynamics in laser ablation plumes in argon atmosphere,” Phys. Plasmas, Vol. 19, pp. 083504 (1-11), 2012.

[8] Z. Zhang and G. Gogos, “Theory of shock wave propagation during laser ablation,” Phys. Rev. B, Vol. 69, pp. 235403 (1–9), 2004.

[9] B. Verhoff, S.S. Harilal, J.R. Freeman, P.K. Diwakar, and A. Hassanein, “Dynamics of femto- and nanosecond laser ablation plumes investigated using optical emission spectroscopy,” J. Appl. Phys., Vol. 112, pp. 093303 (1–9), 2012.

[10] J.M. Fishburn, M.J. Withford, D.W. Coutts, and J.A. Piper, “Study of the fluence dependent interplay between laser induced material removal mechanisms in metals: Vaporization, melt displacement and melt ejection,” Appl. Surf. Sci., Vol. 252, pp. 5182–5188, 2006.

[11] M. Stafe, I. Vladoiu, C. Negutu, and I.M. Popescu, “Experimental investigation of the nanosecond laser ablation rate of aluminum,” Rom. Rep. Phys., Vol. 60, pp. 789–796, 2008.

[12] I. Vladoiu, M. Stafe, C. Negutu, and I.M. Popescu, “Nanopulsed ablation rate of metals dependence on the laser fluence and wavelength in atmospheric air,” U.P.B. Sci. Bull. Series A., Vol. 70, pp. 119–126, 2008.

[13] B.N. Chicbkov, C. Momma, S. Nolte, F. Von Alvensleben, and A. Tiinnermann, “Femtosecond, picosecond and nanosecond laser ablation of solids,” Appl. Phys. A., Vol. 63, pp. 109–115, 1996.

[14] Z. Chen, D. Bleiner, and A. Bogaerts, “Effect of ambient pressure on laser ablation and plume expansion dynamics: A numerical simulation,” J. Appl. Phys., Vol. 99, pp. 063304 (1-9), 2006.

[15] K. Sasaki and N. Takada, “Liquid-phase laser ablation,” Pure Appl. Chem. Vol. 82, pp. 1317–1327, 2010.

[16] B. Kumar, and R. K. Thareja, “Growth of titanium nanoparticles in confined plasma,” Phys. Plasmas, Vol. 19, pp. 033516 (1–7), 2012.

[17] A. Kruusing, “Underwater and water-assisted laser processing: part 2—etching, cutting and rarely used methods,” Opt. Lasers Eng. Vol. 41, pp. 329–352 2004.

[18] S. Zhu, Y.F. Lu, M.H. Hong, and X.Y. Chen, “Laser ablation of solid substrates in water and ambient air,” J. Appl. Phys. Vol. 89, pp. 2400-2403, 2001.

[19] D.N. Patel, R.P. Singh, and R.K. Thareja, “Craters and nanostructures with laser ablation of metal/metal alloy in air and liquid,” Appl. Surf. Sci. Vol. 288, pp. 550-557, 2014.

[20] M. Geiger, W. Becker, T. Rebhan, J. Hutfless, and N. Lutz, “Increase of efficiency for the XeCl excimer laser ablation of ceramics,” Appl. Surf. Sci. Vol. 96-98, pp. 309–315, 1996.

[21] A. Dupont, P. Caminat, P. Bournot, and J.P. Gauchon, “Enhancement of material ablation using 248, 308, 532, 1064 nm laser pulse with a water film on the treated surface,” J. Appl. Phys. Vol. 78, pp. 2022-2028, 1995.

[22] S. Zhu, Y.F.Lu, and M.H. Hong, “Laser ablation of solid substrates in a water-confined environment,” Appl. Phys. Lett. Vol. 79, pp. 1396–1398, 2001.

[23] M.H. Mahdieh, M. Nikbakht, Z. Eghlimi Moghadam, and M. Sobhani, “Crater geometry characterization of Al targets irradiated by single pulse and pulse trains of Nd:YAG laser in ambient air and water," Appl. Surf. Sci., Vol. 256, pp. 1778–1783, 2010.

[24] J.H. Yoo, S.H. Jeong, R. Greif, and R.E. Russo, "Explosive change in crater properties during high power nanosecond laser ablation of silicon," J. Appl. Phys., Vol. 88, pp. 1638-1649, 2000.

[25] S. Palanco, S. Marino, M. Gabás, S. Bijani, L. Ayala, and J. R. Ramos-Barrado, "Micro- and nanoparticle generation during nanosecond laser ablation: correlation between mass and optical emissions," Opt. Express, Vol. 22, pp. 3991–3999, 2014.

Marzieh Akbari Jafarabadi was born in Tehran. She received BSc degree in Atomic and Molecular Physics from Shahid Beheshti University, Tehran, Iran, in 2007. She received her MSc degree in Atomic and Molecular Physics from Iran University of Science and Technology, Tehran, Iran, in 2009. She is presently a PhD student of Atomic and Molecular Physics in Iran University of Science and Technology, Tehran, Iran, since 2011.

Her research interests include laser matter interaction (laser ablation of metals) and laser plasma interaction.

Ms. Akbari is now a member of the Iran Society of Physics and Iran Society of Applied Optics and Photonics.

Mohammad Hossein Mahdieh was born in Tehran. He received BSc and MSc degree in Applied Physics from Sharif University of Technology, Tehran, Iran, in 1988, and 1992, respectively. He graduated from the University of Essex, Essex, UK and received his PhD in Physics (Laser Plasma Interactions) in 1996.

In 1997 he joined Iran University of Science and Technology (IUST), Tehran, Iran, where he is presently an academic staff in the department of physics. His current researches interests include high power pulsed lasers, and their interactions with plasma and matter, and laser propagation in different media.

Professor Mahdieh is a member of the Iran Society of Physics, Iran Society of Applied Optics and Photonics. He also has become a member of the International Advisory Committee of International Symposium on Gas Flow and Chemical Lasers & High Power Lasers GCL&HPL (presently renamed to High Power Laser Systems and Applications) since 2010.