The role of target warming in the synthesis of silicon nanoparticles by picosecond laser ablation in liquid

D. M. Popovic[*], A. A. Zekic[*], M. Trtica[**], J. Stasic[**]

[*]University of Belgrade-Faculty of Physics, Studentski trg 12, 11001 Belgrade, Serbia, dusan@ff.bg.ac.rs, andrijana@ff.bg.ac.rs
[**]Vinca Institute of Nuclear Science, University of Belgrade, P.O. Box 522, 11001 Belgrade, Serbia, etrtica@vinca.rs, jelsta@vinca.rs

ABSTRACT

Silicon-based nanoparticles were produced by irradiating a single crystal silicon target in de-ionized water with Nd:YAG laser at wavelengths of 1064 nm. The additional heating of the target by continuous laser was applied during the experiment and the targets that differ in volume, were used. In the work presented here we further investigated the role of thermal effects, such as warming of the target, on the LAL process.

The noticeable shift of NPs size distribution caused by using the targets of different volumes was reported.
Keywords: silicon nanoparticles; laser ablation in liquid; picosecond laser

1 INTRODUCTION

Silicon nanoparticles (NPs) find applications and potential applications in numerous fields. For example, silicon NPs are important for applications in optoelectronics [1,2]. Aslo, there has been rapidly increasing interest in design and synthesis of siliconbased nanostructured materials for bioapplications [3,4]. In the field of theoretical studies, silicon NPs play important role in the study of fundamental quantum effects [5,6]. The other silicon-based nanoparticles attract attention of authors; silica NPs has possible application for the synthesis of functional materials.

The one of the most promising method for the producing of silicon-based NPs is the laser ablation of solid target in liquid (LAL), because it's relatively simple, fast and "green". The certain extent of the NPs properties tailoring could be accieved by changing the parameters in the LAL process. Also, LAL provides easy material handling because NPs are produced directly in the liquid.

In our previous work, we investigated the LAL process of producing Si NPs with pulsed infrared transverse excited atmospheric (TEA) CO_2 laser and we analyzed the role of thermal effects on the NP's size distribution [7]. In experiments that followed, the main focus of the work was the analyzing of the role of thermal effect on the LAL process. Therefore, the target was additionally heated during the experiment by applying the continuous laser [8].

In the work presented here, in attempt to further investigate the role of thermal effects on the LAL process, the silicon single crystals that differ in volume was used as the target.

2 EXPERIMENTAL METHODS

The silicon single crystal targets that differ in volume were used. The thickness of all used targets (square shape single crystal silicon plate) was the same, while the sizes of their surfaces were $0{,}25cm^2$ and $1{,}96cm^2$. The targets were placed on the bottom of the vessel filled with 5ml of de-ionized water. The water layer thickness was kept to about 3mm during the experiments and the silicon target was fixed at the position during laser irradiation.
The picosecond pulse duration laser (pulse duration 150 ps, pulse energy 7mJ/pulse, repetition rate 10Hz) both with continuous green laser (power about 150mW) was used during the LAL experiment. The laser exposure time was 7 minutes. The additional continuous green laser was applied during the LAL process and for 15 minutes immediately prior to application of pulse laser. The both laser spots, continual and pulsed, were placed at a fixed position in the centre of the silicon target.

The solution obtained by the LAL was dropped onto the silicon substrates and allowed to dry under atmospheric pressure at room temperature. The dried substrates were inspected using a JEOL 840A instrument equipped with an INCA Penta FETx3 EDX microanalyzer (SEM). For EDX analysis the aluminum substrate was used. All SEM/EDX images were recorded within the 24 hours after the laser irradiation of the targets.

3 RESULTS AND DISCUSSION

The main goal of our work presented here was to enlight further the role of warming of the target on the LAL process. To acchieve this goal, we provide additional heating of the target surface by applying the additional continuous laser during the experiment and we used the targets that differs in volume. In attempt to minimize the heating of the water layer, we used the green laser.

The use of picosecond pulse duration in our experiment provides that the nonthermal photon-based ablation could be neglected [9].

Figure 1 shows the SEM micrograph image of silicon nanoparticles prepared by the picosecond laser ablation in de-ionized water for the size of target surface (a) 1,96cm^2 and (b) 0,25cm^2. The continual laser was applied immediately prior and during LAL process in both experiments.

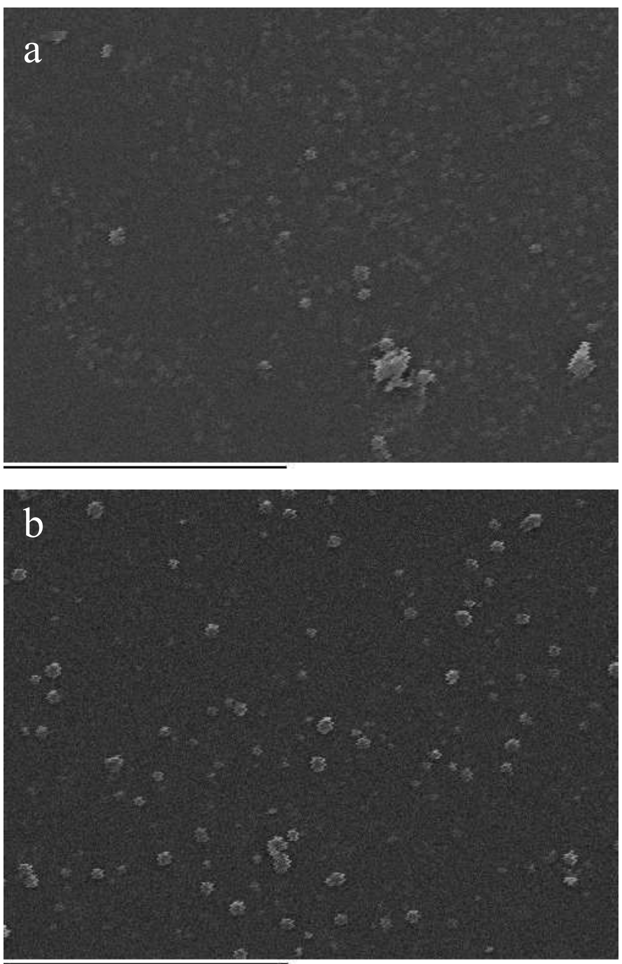
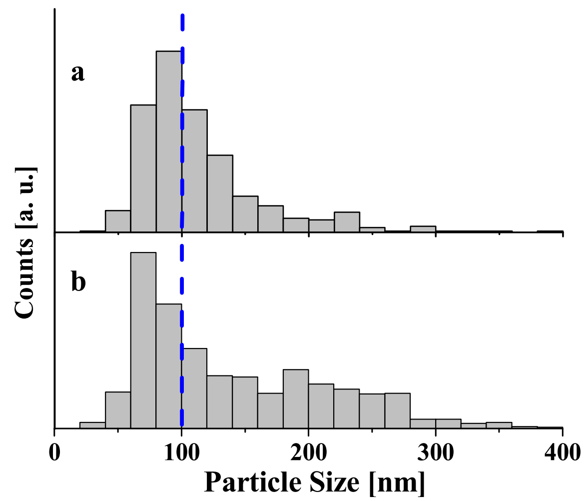


Figure 1: SEM micrograph image of silicon nanoparticles prepared by the picosecond laser ablation in de-ionized water for the size of target surface (a) 1,96cm^2 and (b) 0,25cm^2 (bars denote 5μm).

The size distribution of produced particles obtained by counting approximately 800 particles in SEM image, are shown in Fig. 2a and 2b, respectively. We considered only particles in the 50nm–400nm size range to achieve a size distribution due to the fact that larger ones were uncommon. All particles taken into consideration have a spherical or a nearly spherical shape. It could be noticed from Fig. 2 that using the different size of the targets changes the NPs size distribution.

In the case of using the smaller target the region under 100 nm was more populated than in the case when the larger target was used. By comparing these results with those we reported earlier [8], it can be noticed that the applying of the continual laser during the LAL process has the similar effect as the use of smaller targets. We suppose that changing the target size as well as applying the continual laser during LAL process change the temperature of the target and therefore change the NPs size distribution.

Because of plasma plume appears over the laser spot on the target surface the chemical composition of the produced nanoparticles will be of silicon oxide. This was confirmed by EDX measurements of produced nanoparticles, but no precise conclusion regards stoichiometry of silicon oxide could be reached.



.

Figure 2 Size distributions of silicon nanoparticles prepared by the picosecond laser ablation in de-ionized water for the size of target surface (a) 1,96cm^2 and (b) 0,25cm^2 (vertical line at 100nm should facilitate comparison).

4 CONCLUSION

In this paper, we studied the production of silicon-based nanoparticles by irradiating the single crystal silicon target in de-ionized water with Nd:YAG laser at wavelengths of 1064 nm and pulse power of 7 mJ/pulse. To provide additional heating of the target surface the additional continual green laser was applied during and immediately prior the ablation process. The silicon single crystals that differ in volume was used as the targets. The shift of NPs size distribution caused by using the different size of the targets was reported. Further investigation in this field could additionally enlighten the role that the temperature of the target plays in the LAL process.

REFERENCES

[1] L. Pavesi, L. Dal Negro, C. Mazzoleni, G. Franco, and F. Priolo, Nature 408, 440, 2000.

[2] R. J. Walters, G. I. Bourianoff, and H. A. Atwater, Nature Mater. 4, 143, 2005.

[3] Y. He, C. Fan, S.-T. Lee, Nano Today 5, 282, 2010.

[4] J.-H. Park, L. Gu, G. von Maltzahn, E. Ruoslahti, S. N. Bhatia and M. J. Sailor, Nature Materials 8(4), 2009.

[5] C. Meier, A. Gondorf, S. Luttjohann, A. Lorke, and H. Wiggers, J. Appl. Phys. **101**, 103112, 2007.

[6] P. Mishra and K. P. Jain, Mat. Sci. Eng. B **95** (3), 202, 2002.

[7] D. M. Popovic, J. S. Chai, A. A. Zekic, M. Trtica, M. Momcilovic and S. Maletic, Laser Phys. Lett. 10, 026001, 2013.

[8] D. M. Popovic, A. A. Zekic, M. Trtica, J. Stasic, NSTI-Nanotech, June 18-21, 2012, Santa Clara, CA, 2, 510, 2012.

[9] S. K. Sundraram and E. Mazur, Nature Materials, 1, 2002.