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# Morphological, elemental and hardness analysis of femtosecond laser irradiated Al targets

Umm-i-Kalsoom [a,b,c], Nisar Ali [b,c,d,*], Shazia Bashir [c], Narjis Begum [e], M. Shahid Rafique [f], Wolfgang Husinsky [b]

[a] Department of Basic Sciences and Humanities, University of Engineering and Technology KSK Campus, Lahore, Pakistan
[b] Institute for Applied Physics, Vienna University of Technology, Vienna, Austria
[c] Centre for Advanced Studies in Physics, GC University, Lahore, Pakistan
[d] Department of Basic Sciences and Humanities, University of Engineering and Technology Lahore, Faisalabad Campus, Fasalabad, Pakistan
[e] Department of Physics, Riphah International University, Islamabad, Pakistan
[f] Department of Physics, University of Engineering and Technology Lahore, Pakistan

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Aluminum (Al) samples were irradiated with femtosecond (fs) laser pulses for different laser fluences under two different environments of vacuum & Oxygen ($O_2$). Nano/Micro structures on the surface of irradiated Al were explored by using Scanning Electron Microscope (SEM). Self-organized patterns like ripples, cellular structures, cluster of particles and cavities are observed by SEM analysis. Single and multiple (100) shot, ablation threshold value and incubation coefficient were also calculated by means of SEM images, under both ambient conditions i.e. vacuum and $O_2$ environments. While the comparison for single and multiple (100) shots shows a decrease in the value of ablation threshold with increase of number of laser pulses due to presence of incubation effect. X-ray Diffraction (XRD) and Energy Dispersive X-ray Spectroscopy (EDX) analysis were utilized for identification of phases and the chemical composition of ablated targets, respectively. A variation in Al content as well as in peak intensities of almost all phases is observed under vacuum treatment. In case of treatment in $O_2$ ambient, oxides of Al are achieved with significantly enhanced concentration of $O_2$. After ablation under vacuum condition reduction in atomic O content (already present on un-ablated target) is observed. The reduced ablation threshold of metals makes the laser material processing mechanisms like cutting, drilling, welding and surface modifications, more effective. The observed reduced ablation threshold of Al in $O_2$ as compared to vacuum, as well as oxidation of Al along with the growth of surface structures make this metal more useful for various industrial as well as scientific applications. Nano-hardness measurement shows an increase in nano-hardness of irradiated targets as compared to un-irradiated ones with the increase of fluence under both ambient conditions.



## 1. Introduction

Femtosecond laser material processing is an important tool due to minimum thermal effects. By irradiating targets to ultrashort pulses, various features like ripples [1–4], hillocks, bumps, cellular structures, cluster of particles and cavities [5–7], can be formed. Laser parameters and environmental conditions play vital role for the formation of these structures [8,9]. The formation of these structures is explainable on the basis of both thermal and nonthermal mechanisms including plasma as well as parametric instabilities, optical and interference effects, surface plasmons, surface tension gradients and Coulomb's explosion [10].

Aluminum (Al) metal, due to its promising properties like good corrosion resistance, low density and good thermal conductivity is a potential candidature for wide range applications in industry, space sciences and electronics, is therefore selected as a target material. Whereas, the tribological properties of the Al surface are not so good and need to be improved for the best usage in wearing environments. This can be done by oxidation of metal surface along with the formation of surface structures. One well known method for oxidation is to ablate the surface of target by laser pulses in the presence of $O_2$ environment [11]. This is one of most exciting motivation of present work.

* Corresponding author at: Department of Basic Sciences and Humanities, University of Engineering and Technology Lahore, Faisalabad Campus, Faisalabad, Pakistan.

*E-mail address:* chnisarali@gmail.com (N. Ali).

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The growth of different surface structures on metals can enhance their various properties e.g. optical absorption [12,13], thermionic properties, photoelectron emission [14] and super-hydrophobicity [15]. The metals with improved properties are useful for various industrial, physical, biological and chemical applications [16].

Significant work is performed on the laser ablation and laser induced structuring of Al. Kikuchi et al. (2001) [17] studied the modification in the surface physical and electrochemical properties of Al after laser irradiation in chemical solution. After getting oxidized the Al targets become beneficial in the field of process industry. Davenport et al. (2005) [18] reported enhancement in the corrosion properties of Al alloys after laser treatment, which make them useful for aerospace applications.

Mannion et al. [19] calculated incubation coefficient of metallic targets after ablating them in air by fs lasers for different number of pulses. From their results it was verified that the ablation threshold value basically depends on the number of pulses applied to the irradiated target.

Gamaly et al. [20] studied the effect of ambient environments (Vacuum & air) on the ablation threshold of irradiated targets of Al, Cu, Fe and Pb. They reported that ablation threshold of metallic targets in air, is half times less as compared to vacuum condition.

Formation of ripples on Al, Si, $CaF_2$ and CR-39 after irradiation with Ti: Sapphire laser has been reported by Bashir et. al. [21]. For all materials nano-ripples with different periodicities are observed. Whereas, on the surface of Al and Si nano-ripples as well as micro-ripples having periodicity of about 1–2 μm have also been revealed. The fluence value for micro-ripple formation is higher than for nano-ripple-formation [21].

The current research work deals with the evaluation of the single and multiple-pulse ablation thresholds of Al as well as incubation coefficients for different ambient conditions (Vacuum, $O_2$) which were explored by SEM analysis. The variations in surface morphology with the variation of fluence, shot number (single and multiple (100 shots)) and ambient environment are also investigated by using SEM analysis. The chemical and phase analysis is performed by using EDX and XRD analysis. A correlation between the growth of surface structures, chemical analyses and generation of stresses under both ambient environments is established. It is observed that the fs laser pulses produce a large variety of nano/micro-structures i.e., LIPSS formation, cellular structures, grains and cavities. The present work gives comprehensive insight of physical processes and mechanisms responsible, for defining ablation threshold, incubation effect and surface structuring. The second motivation behind this work is that very rare work is reported regarding ablation of Al in Oxygen. Most of the reported research work is performed in air and vacuum. The laser ablation of Al in $O_2$ environment is responsible for enhanced energy deposition to the target due to enhanced energy coupling mechanisms and oxygen assisted exothermic reactions [22]. This will cause reduction in ablation threshold for both single and multiple pulses and hence can make laser-material processing (cutting, drilling, welding and surface modifications) more efficient tool in the industry.

## 2. Experimental setup

Square shaped Al targets with dimensions of $10 \times 10 \times 5\ mm^3$ were grinded, polished and ultrasonically cleaned for 30 min, prior to irradiation experiments. The polished targets were placed inside the vacuum chamber, which was evacuated to $10^{-3}$ mbar, pressure.

Chirped Pulse Amplification (CPA) Ti: sapphire laser (repetition rate 1 KHz, wavelength 800 nm, pulse duration 30 fs), was used to irradiate the Al targets inside the vacuum chamber. The unfocused beam spot size was about 1.5 cm whereas the average value of spot size on sample surface is 83 μm after focusing through 20 cm focal length lens.

The irradiation experiments were performed in two steps.

1. The initial set of experiments was performed for the evaluation of ablation threshold and incubation coefficients, of Al for single pulse irradiation at 10 different energies of 20, 30, 40, 50, 60, 70, 80, 130, 180 and 230 μJ with corresponding fluences of 0.19, 0.28, 0.37, 0.47, 0.56, 0.65, 0.74, 1.21, 1.67 and 2.14 J $cm^{-2}$, respectively.

1- In the former experiments the Al targets were irradiated with 100 pulses of fsec lasers for different fluences, 0.37, 0.56, 1.21, and 2.14 J $cm^{-2}$. This data is presented in SEM results.

These two set of experiments were executed under vacuum condition and in $O_2$ ambient at 133 mbar, pressure.

Surface topography of irradiated Al targets was performed using SEM (FEI-QUANTA 200F, Netherlands) analysis. To evaluate chemical composition, phases, crystallinity and strain/stress variation EDX and XRD (X'Pert PRO (MPD)) analysis were employed, respectively.

## 3. Results and Discussions

### 3.1. SEM Analysis

SEM images of Fig. 1(a)–(d) illustrate the surface morphology of Al after irradiation with 100 laser pulses at various fluence of (a) 0.37, (b) 0.56, (c) 1.21 and (d) 2.14 J $cm^{-2}$, when treated under vacuum. For minimum fluence value (0.37 J $cm^{-2}$) lamellar structures (nano-ripples) capped with nano-protrusions, cavities and spherical tips are shown in Fig. 1(a). With an enhancement in fluence value to 1.21 J $cm^{-2,}$ the nano- protrusions and nano-cavities, become distinct with an appearance of cellular structures and nano-rims. The nano-LIPSS formation can be related to the presence of incubation effect that is the basic cause for the defect generation and production of plasmonic effects. These plasmonic effects change the dielectric function of the material, thus alters the laser intensity distribution, hence create the maxima and minima of intensity and cause the formation of nano-LIPSS [23,24].

The mechanism behind formation of nano-ripples is excitation of plasmons that generate periodic augmentation in local fields of surface layers. When the fs laser beam in ultrashort time scale is made to fall on the surface of target, abundant electrons become excited, causing generation, of high density plasmas. This will cause the alteration of surface physical and optical properties. The change in optical properties will modulate the electromagnetic intensity distribution and will affect the laser matter interaction phenomenon intensively. This intensity distribution will persuade the localized strong fields causing the phenomenon of Coulomb's Explosion (CE) at nanoscale. Which is the basic reason behind the generation of nano LIPSS [21,25].

The cavities and protrusions are developed randomly over the laser ablated region after laser irradiation due to in-homogenous energy deposition, caused by the enhanced surface roughness [26]. The initial pulses introduce surface roughness and enhance it [27]. While the remaining pulses causes the structuring and variation in the surface structures that is related to the in-homogenous energy deposition. The surface roughness, presence of impurities, voids, in-homogeneities and non-uniformities on the surface of Al are also responsible for non-uniform laser energy absorption. This variation in energy can be described on the basis of spatial