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# Laser shock cleaning of radioactive particulates from glass surface



<sup>a</sup> Advanced Fuel Fabrication Facility, Bhabha Atomic Research Centre, Tarapur 401504, Maharashtra, India <sup>b</sup> Laser & Plasma Technology Division, Bhabha Atomic Research Centre, Mumbai 400085, Trombay, India

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### ABSTRACT

Efficient removal of Uranium-di-oxide (UO<sub>2</sub>) particulates from glass surface was achieved by Nd–YAG laser induced airborne plasma shock waves. The velocity of the generated shock wave was measured by employing the photo-acoustic probe deflection method. Experiments were carried out to study the effect of laser pulse energy, number of laser exposures and the separation between the substrate surface and the onset point of the shock wave on the de-contamination efficiency. The efficacy of the process was estimated monitoring the alpha activity of the samples before and after laser shock cleaning using a ZnS (Ag) scintillation detector. Significant cleaning efficiency could be achieved when the substrate was exposed to multiple laser shocks that could be further improved by geometrically confining the plasma. No visual damage or loss in optical quality was observed when the shock cleaning was found to be significantly larger than that possible by conventional laser cleaning. Theoretical estimate of the shock force generated has been found to exceed the van der Waal's binding force for spherical contaminant particulate.

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## 1. Introduction

The use of lasers in surface cleaning for removing particulate contamination is a modern technology and has major advantages over the conventional wet cleaning methods, e.g., environmental friendliness, dry nature of cleaning that generates very little secondary waste, non-contact process, selective removal of the contaminants, be it particulates or extended layers without affecting the substrate and possibility of automated systems with minimum manual intervention. These advantages have resulted in employing laser based surface cleaning technology in many areas of engineering and science, e.g. nuclear industry [1–3], restoration of art works [4–6], semi-conductor industry [7–9] etc. In case of loose contamination that is bound to the substrate surface predominantly by van der Waal's force [10], exposure to a laser pulse of suitable wave length and fluence results in absorption of the incident energy by the substrate or the contaminants causing their expansion due to rapid rise in temperature. If the force experienced by the particulate as a result of this thermal stress exceeds the adhesion force, the particulates are dislodged from the substrate. However, direct exposure of the substrate to the intense laser beam may cause permanent damage to the

substrate, especially if the substrate is brittle or has low melting point. As an alternative to the thermal stress induced cleaning, laser shock cleaning is a promising technique that allows removal of small particulates from the substrate surface without requiring it to be exposed directly to the laser beam. The shock wave is generated by focusing the laser beam at a specific distance above the surface to be cleaned in a gaseous environment. This results in dielectric breakdown and ionization of the medium generating rapidly expanding plasma at the point of focus. This results in the formation of a shock wave which moves outwardly at supersonic velocity. The resulting drag force acting on the particulates, if exceeds the van der Waal's binding force, can result in their expulsion. There exist many reports on the removal of contaminants, of size varying from micro-meters to tens of nano-meters, from a variety of substrate surfaces employing the method of laser shock cleaning [11–13]. Apart from the fact that the substrate surface remains unharmed, the laser shock cleaning process can work effectively for both metallic and dielectric substrates and particulates, irrespective of the operating wavelength.

Pulsed laser assisted removal of loose radioactive contaminants from metallic surfaces has generated considerable interest mainly because this process produces very little secondary waste and also reduces the possibility of personnel exposure. Although several papers report on the removal of radioactive oxide layers [14,15] and particulate [16–18] contamination from metallic surfaces using lasers, reports on removal of radioactive contaminants from

<sup>\*</sup> Corresponding author. Tel.: +91 2525 244165; fax: +91 2525 244913. *E-mail addresses*: nontee65@rediffmail.com, nontee65@yahoo.com (A. Kumar).

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dielectric surfaces are limited [19]. Removal of such contamination off dielectric surfaces assumes great significance in the nuclear industry where glove boxes, which house common radio toxic materials like uranium, plutonium etc. and acquire loose contamination on their inner surfaces over a period of usage, are basically constructed using large glass panels. Work has been carried out in the past to clean contaminated glass samples utilising lasers although with a limited success. Masahiro et al. [20] used CO<sub>2</sub>, N<sub>2</sub> and Nd-YAG lasers to clean marked black stains on glass surface. While satisfactory cleaning could be observed with CO<sub>2</sub> or N<sub>2</sub> lasers, usage of YAG laser resulted in cracking of the glass surface. Cleaning of silica glass surfaces contaminated with alumina particles by a KrF laser resulted in systemic damage in the form of pits on the surface [21]. Hua et al. [22] removed micron sized silicon-di-oxide particulates from a smooth glass surface by vaporizing a thin paint film pre-coated on the surface using a YAG laser. Pleasants et al. [23] and Shukla et al. [24] achieved cleaning of alumina and silica particles from glass and silica substrates utilising a KrF excimer laser and TEA CO<sub>2</sub> laser, respectively. It is, however, generally evident from the literature that the process of cleaning of glass by directly exposing it to the laser beam makes it susceptible to damage unless the laser parameters are carefully adjusted. On the other hand, laser shock cleaning, that does not require any interaction between the laser beam and the substrate could be a viable alternative method of cleaning contaminated glasses with minimal possibility of substrate damage.

In this paper we report the results of our studies on cleaning of  $UO_2$  particulates off glass surface by shock waves induced by a Q-switched Nd–YAG laser capable of generating pulses of 6 ns duration. Decontamination efficiency, defined as the percentage of initial activity removed, was evaluated by counting the alpha activity of the samples before and after laser exposure using a ZnS(Ag) scintillation detector. The laser treated glass surface was analyzed for any possible surface damage by optical microscopy and spectrophotometry.

#### 2. Experimental

In the first set of experiments the velocity of the shock waves as a function of laser energy at varying distances from the focal spot was estimated. To this end, a Q switched Nd–YAG laser operating at 1064 nm and capable of delivering a maximum multi-mode energy of 1.6 J over a beam area of ~1 cm<sup>2</sup> and pulse duration of 6 ns was used in conjunction with a 3 mW He–Ne laser operating at 632 nm. Fig. 1 shows the schematic diagram of the experimental set up. The laser shock was produced by focussing the Nd–YAG laser beam (propagating in a horizontal plane) using an antire-flection coated BK7 plano-convex lens of focal length 100 mm. The spot size was 1.2 mm, which corresponds to a power density of  $1.4 \times 10^9$  W/cm<sup>2</sup>. The energy of the laser pulse was measured using a wedge plate in conjunction with a pyro-electric Joule meter as



Fig. 1. Schematic of the experimental set up for determination of shock wave velocity.

shown in the figure. The He–Ne beam, also in the horizontal plane but orthogonal to the Nd–YAG laser beam, was used to study the shock wave propagation using the beam deflection technique as discussed below.

The scanning of the probe He-Ne beam below the focal point in the vertical direction allowed probing the shock velocities at different locations. Deflection of the probe beam due to the gradient of refractive index induced by the passage of the shock wave was detected by using a sensitive fast detector (Thorlab: DET10A) coupled to a Tektronix digital storage oscilloscope. A narrow band pass filter (central wave length 633 nm) and a dichroic mirror that completely reflects the 1064 nm were placed in front of the detector to avoid the presence of any stray radiation. The probe beam was made to scan the region below the focal plane in steps of 1 mm and for each position data was acquired with varying pulse energies. The temporal evolution of the beam deflection provided the time of flight of the shock front measured at different positions with respect to the YAG laser focal spot. Fig. 2 shows the beam deflection signals for three different laser pulse energies for which the probe beam was at 7 mm away from the YAG laser focal spot. Shock wave velocity at different positions of the probe beam was calculated from the acquired time of flight data from the deflection signals. The shock pressure was then estimated using the Rankine-Hugoniot equations [25].

In the next set of experiments, the shock wave induced cleaning efficiency was studied with glass substrates contaminated with  $UO_2$  particulates as a function of energy of the laser pulse for single and multiple exposures in steps of 1 mm vertically down the focal point (Fig. 3). The experimental conditions and set up were basically the same used for determination of velocity of shock waves except the He–Ne laser, photo- detector and other optical components used therein.

Glass plates measuring  $15 \text{ mm} \times 15 \text{ mm} \times 3 \text{ mm}$ , cleaned in demineralised water and acetone to remove any contamination present on the surface, were used as the samples. Minute quantity of UO<sub>2</sub> powder dispersed in a small volume of iso-propyl alcohol was sonicated in an ultrasonic bath for fifteen minutes. A droplet of the prepared suspension was deposited over a central spot of diameter 8–10 mm of the glass sample and was allowed to dry in air. The alpha activity of the prepared samples was measured using a ZnS (Ag) scintillation detector and samples with activity lying within  $\pm 2.5\%$  were chosen for the experiment. The activity of the UO<sub>2</sub> samples was  $\sim 10 \text{ Bq/cm}^2$ . The minimum detectable activity (MDA) of the scintillation detector was  $\sim 12$  disintegrations per minute (dpm). The



Fig. 2. He-Ne laser beam deflection signals for three different laser shock pulse energies. Laser pulse energy (from top): 600 mJ, 800 mJ and 1000 mJ.

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