LASER ASSISTED SURFACE CLEANING: A STUDY VIS-A-VIS INDIAN NUCLEAR FUEL CYCLE

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Abstract

Occupational exposure to radiation occurs as a result of work associated with different stages of nuclear fuel cycle from mining to de-commissioning. The essential objective of radiation protection is to protect human health against the effects of radiation resulting from handling of radioactive materials. Although adoption of appropriate design of handling equipments and operational procedures offers protection but it cannot provide complete safeguard as the conventional methods often involve personnel coming into contact of the radioactive surfaces mainly with jobs involving cleaning of contaminated surfaces. To this end laser has proved to be an ideal tool as the entire cleaning operation with laser can be performed in a remote manner thus greatly reducing the possibility of exposure to the personnel. Further, the laser parameters can be carefully controlled to dramatically minimize the secondary wastes. No wonder that laser assisted surface cleaning has decided advantage as a decontamination tool when employed in nuclear industry. It has the potential to remove both loose and fixed contaminations from metallic as well as dielectric substrates without causing any harm to the surface underneath. This paper provides an insight to our work of employing a laser in multiple ways to affect surface cleaning in nuclear industry.

Introduction

India is persuading a three stage nuclear power programme to meet its future energy needs. This three stage program is based on a closed fuel cycle, wherein the spent fuel of one stage is reprocessed to produce the fuel for the next stage. The first stage of this programme involves using the natural uranium in Pressurized Heavy Water Reactors (PHWR’s). The plutonium that is recovered from the reprocessing of the spent nuclear fuel from the first stage along with the non-fissile depleted uranium and thorium make the fuel for the second stage that essentially is a fast breeder reactor. The depleted uranium and thorium can breed additional fissile material here viz., plutonium and uranium-233 respectively.

The third stage is based on the operation of thorium and uranium-233 fuelled nuclear reactors. Thus, fabrication of Plutonium and U-233 bearing MOX fuel and reprocessing of spent fuel plays a major role for successful implementation of the three stage nuclear power programme.
The major hazard in a plutonium / U-233 handling facility e.g. in a fuel reprocessing plant or a fuel fabrication plant arises from the possibility of inhalation or ingestion of the radio-toxic material by personnel or from the exposure of radiation caused primarily by gamma rays and neutrons. Storage and treatment of highly active alpha wastes is also another concern for these facilities. Thus plant design, equipment design and process flow sheet for these facilities should aim for minimising both manrem expenditure and generation of wastes. The high radio-toxicity and large biological half life associated with plutonium necessitates installation of all processing equipments inside leak tight glove boxes and heavily shielded glove boxes or hot cells for U-233 owing to the hard $\gamma$ activity of daughter products of U-232 with remote operation and automated facilities.

In a MOX fuel fabrication plant, fuel pins are fabricated by loading fuel pellets, fabricated by conventional powder-pellet route using powder metallurgy techniques followed by encapsulation with a suitable technique. The pellet loading operation involving pushing the fuel pellets into the fuel tube, evacuation process during top end plug welding and in general handling the tubes inside glove boxes results in the presence of loose oxide particulates contamination on its surface, in general, and near the edges in particular. The loose contamination present on the outer surface of the fabricated fuel pins needs to be removed before their removal from the glove box for further processing and assembly. Conventional methods based on mechanical and chemical techniques are not suitable for decontamination of thin walled fuel pins as they are abrasive in nature leading to the possibility of clad damage. The manual method of decontaminating the fuel pins is most common and involves cleaning the pin surface inside the glove box with soft wet cloth and repeating the procedure after their removal from the glove box until the permissible level of activity is reached. This process is time consuming, leads to undue exposure of the personnel, generates large alpha active secondary waste and increases the possibility of air borne activity in the working area. Ultrasonic cleaning, an alternate technique currently adopted in many industrial facilities world-wide, requires submerging of the fuel pins in a specially designed water filled tank and subjecting them to ultra-sonic agitation. Although ultrasonic decontamination results in the dislodging of larger size loose particulates, it also generates large quantity of liquid waste necessitating an additional step of treating the active liquid for re-use / disposal.

Lasers can play an important role in surface decontamination of nuclear fuel pins as the process can be performed remotely with minimum generation of secondary wastes.

As discussed earlier, nuclear fuel fabrication plants and reprocessing plants handle bulk of plutonium material either in liquid form or in powder form inside glove boxes. The entire internal surfaces of the glove boxes get severely contaminated during handling of plutonium, more severely in the case where PuO$_2$ is handled as fine powder. These glove boxes need either renovation or disposal after several years of service primarily due to the build up of activity to an alarming level and deteriorated internal conditions. In case of disposal, the activity of the glove box needs to be reduced considerably so as to treat it as a low level waste that, in turn, reduces the disposal cost. On the other hand, the cleaning of the internal surfaces of the glove box will allow the renovation work to be performed at a lower radiation field. Conventionally the internal surfaces of the glove boxes are cleaned using strippable gels or chemicals. In the former case, the
gel is first applied to the contaminated surfaces and is allowed to cure. Later the layer so formed is peeled from the surface and stored as a solid waste. In the chemical process complex blends of acids and other chemicals are used manually in a multi-step process. There is prospect of cleaning the contamination from the inner surface of the glove box by locating the laser outside as there is enough literature to suggest that reverse cleaning is a possibility. Wherever the contamination is on the surface and fixed in nature the generated secondary waste can be greatly reduced by employing laser as the cleaning agent. This assumes larger significance as many reactors of the country are ageing and the prospect of decommissioning looms ahead. For recycling of precious materials and reduction of waste and exposure to personnel laser surface cleaning can be an efficient tool.

There are fundamentally three different approaches for laser assisted surface cleaning for removing particulate contamination from a substrate surface- 1) Dry laser cleaning, 2) Steam laser cleaning and 3) Laser shock cleaning. The dry laser cleaning involves direct interaction of the laser beam with either the contaminants or the surface or both. The micron /sub-micron sized particulates are adhered to the substrate by short range attractive forces e.g., Van der Waal’s force that exists between both polar and non-polar substances. The magnitude of the adhesion force depends on the nature of the particle; its size, shape and contact area with the substrate. Absorption of energy from laser pulse either by the particulate or the substrate or by both can result in the rapid rise of temperature leading to the generation of a thermo-elastic force. The particulates can get dislodged from the substrate when the value of the generated force is more than the adhesion force. The steam cleaning approach is normally associated with the application of a thin liquid film on the surface prior to the interaction of the laser beam. The thrust generated by the explosive vaporisation of the liquid film under the action of the laser pulse dislodges the particulates from the surface. In laser shock cleaning, a shock wave is generated by focusing the laser beam at a specific distance above the surface to be cleaned in a gaseous or liquid environment. High electric field at the focal point 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.

In Advanced Fuel Fabrication Facility, Tarapur we have carried out experiments for decontamination of metallic and dielectric surfaces using dry laser cleaning and laser shock cleaning techniques. The dry laser cleaning technique has been successfully implemented in the decontamination of PFBR fuel pins. The following paragraphs will elaborate some of our experimental work and their results.

**Experimental and results**

**Dry laser forward cleaning: Decontamination of PFBR fuel pins**

The second stage of India’s nuclear power programme is based on Fast Breeder Reactors. A Prototype Fast Breeder Reactor (PFBR) is under construction at Kalpakkam, Tamilnadu. The reactor is based on Uranium-Plutonium Mixed Oxide (MOX) fuel, 20% cold worked D9 stainless steel clad tube and liquid sodium coolant. Fabrication of fuel pins for this reactor is now
in progress at Advanced Fuel Fabrication Facility, Tarapur. Decontamination of fuel pins was carried out using a Q-switched Nd-YAG laser operating at 1.06 µm and capable of delivering a maximum energy of 1.6 J over a pulse of duration 6 ns (FWHM). The laser emits a multimode beam of cross section ~ 1 cm². A schematic diagram of the experimental set-up is shown in Figure 1. A work station capable of providing simultaneous rotational and translational motion to the fuel pin was made use of for this work and was installed inside a specially built fume-hood. The far end of the pin was held in a chuck, mounted on the shaft of the rotational stage. Proper supports were provided to hold the 2.60 meter long fuel pin horizontally. The linear and translational motions were effected through a programmable controller. The laser beam was steered into the fume hood through a narrow opening by means of appropriately arranged mirrors. A suction mechanism ensured that the ejected particulates from the interaction zone found their way into the HEPA filter attached with it. This mechanism along with the appropriate pressure gradient maintained inside the fume hood ensured that no airborne activity contaminated the working area. Further, usage of an inert purge gas prevents oxidation of the clad surface at the interaction zone as well as re-deposition of the ejected particulates. The pitch of rotation of the workstation, and the repetition rate of the laser were so adjusted as to irradiate

![Schematic Diagram](image)

**Fig. 1: Schematic of PFBR fuel pin decontamination setup**

the entire active area of the fuel pin surface by the stationary laser beam with marginal overlap between the successive exposures. A number of precursory experiments on small contaminated samples were carried out to ascertain the laser parameters required to bring down the activity to the permissible level [1]. It was observed that exposing the sample surface to ~ eight laser pulses with a fluence value of ~700 mJ/cm² @1064nm could bring down the contamination level to the acceptable level. Following the optimization of laser parameters, laser assisted decontamination of the PFBR fuel pins was carried out. Figure 2 shows the activity per unit area (Bq/cm²) of hundred fuel pins before and after laser decontamination. It is seen that laser cleaning could always bring down the activity significantly ensuring the removal of the loosely bound
contaminants from the fuel pin surface. Decontamination factor (ratio of the initial to the final activity) as high as $10^4$ was achieved. The effect of laser exposure on the clad surface was then evaluated by carrying out SEM, EPMA and micro-hardness analysis. It was observed that this process does not alter the surface morphology and mechanical properties of the clad tube. Being a dry and non contact process, generation of solid waste and personnel exposure to radiation was also reduced. Figure 3 shows a typical comparison of radiation dose received by an individual radiation worker while carrying out decontamination of ten representative fuel pins by three different methods. It is observed that use of laser as the decontamination tool resulted in minimum exposure.

![Fig. 2: Activity of hundred PFBR fuel pins before and after laser decontamination](image)

![Fig. 3: Comparison of radiation exposure for three different methods of decontamination](image)
Dry laser reverse cleaning - Cleaning of contaminated laminated glass

Our experiments were conducted on laminated glass pieces of size 2 inch x 2 inch and thickness ~ 6 mm. One of the clear surfaces of these glasses was contaminated by smearing UO$_2$ powder on it. A pulsed Nd-YAG laser capable of delivering a 6 ns pulse at 1064 nm and 532 nm wavelengths was used as the cleaning tool. The sample was scanned manually to clean the entire area. To estimate the decontamination efficiency, which is defined as the percentage of initial activity removed, alpha activity of the samples was measured before and after laser irradiation by making use of a ZnS(Ag) scintillation detector. The float glass is basically a soda lime glass which exhibits maximum transmission at @500nm wavelength.

Experimentally we found that the laminated glass slab used by us exhibits a maximum transmission of ~ 85% at 532 nm wave length and ~ 50 % for 1064 nm. Most of the earlier works on cleaning of contamination from glass surfaces have made use of UV radiation derived from either ArF or XeCl lasers. However for laminated glass panel cleaning, UV light cannot be used because the intermediate PVB layer blocks the transmission of UV photons through it. In our experiment we have irradiated the UO$_2$ contamination from the back side of the glass as the magnitude of the generated thermo elastic force will be more here in comparison to front side exposure. Practically too, it is an easy option as the laser beam in that case can be readily directed towards the glass panel. Figure 4 shows the percentage of decontamination efficiency as a function of fluence obtained after single pulse exposure.

![Graph showing variation of single pulse decontamination efficiency with fluence](image)

**Fig. 4: Variation of single pulse decontamination efficiency with fluence**

It is seen that very efficient cleaning at much lower fluence can be obtained with 532 nm wave length because of its better transmission through the laminated glass sheet as well as better absorption in UO$_2$. Although we have calculated DE after single pulse exposure, its value may
increase with more number of exposures for the fluence values used here. We have not observed any visual defects in the form of cracks or any marks on the glass surface as a result of the laser irradiation. Even though we have carried out initial experiments with UO$_2$ powder, this method of cleaning can be used for plutonium powders as well with an appropriate change in the fluence values. For practical application of this method, lasers with beam delivery through articulated arm will be preferred. Manually the beam can be scanned over the surface. For collection of dislodged particulates from the interaction zone the ventilation system of the glove box itself can be made use of, although, with an increased number of air changes. Other than renovation or disposal, cleaning of the inner surfaces of the glove boxes will increase the life of the glove boxes in addition to reducing the exposure to laboratory personnel.

**Laser shock cleaning of contaminated samples**

In dry laser cleaning, the contaminants are removed by exposing the contaminated substrate directly with a laser beam. 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 or liquid 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 outward 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. Experiments on cleaning of UO$_2$ particulates off metallic surface was carried out with shock waves induced by a Q-switched Nd-YAG laser (2$^{nd}$ harmonic @532 nm) capable of generating pulses of 6 ns duration in air and water. 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 samples were analyzed for any possible surface damage by optical and electron microscopy. 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 by beam deflection technique [2]. The shock pressure was then estimated using the Rankine-Hugoniot equations. The shock velocity was found to be varying between 2000 m/s to 800 m/s in air and between 3,500 m/s to 1800 m/s in water as the distance from the focal spot changed from 0.7mm to 2 mm respectively. The peak pressure of the shock wave at a distance of 0.7 mm was found to be ~ 3 MPa in air and ~ 3.5GPa in water and then decayed exponentially with increase in gap distance. Figure 5a to 5d shows the variation in decontamination efficiency (DE) obtained in water (5a and 5b) and in air (5c and 5d), as a function of laser pulse energy and number of exposures for two different orientations of the sample wrt the focal point i.e placing the sample horizontally and vertically at the same distance (0.7mm) from the laser focal spot. DE was seen to increase with increase in laser pulse energy and the number of exposures Under identical conditions of exposure DE was found to be always more for the samples treated holding vertically in comparison to the samples treated placing horizontally. The reduced DE in case of horizontally held samples was due to re-deposition of
the some of the removed particulates. Re-deposition was more in water than air due to higher density of water resulting inefficient cleaning in water particularly with laser pulses of lower energy and less number of exposures (Fig.5a).

![3D graph showing variation of DE with laser pulse energy and number of exposures.](image)

**Fig. 5**: Variation of DE with laser pulse energy and number of exposure. a) Sample placed horizontally in water, b) sample placed vertically in water, c) sample placed horizontally in air, d) sample placed vertically in air.

Most efficient cleaning was obtained when the samples were treated in water and was held vertically. DE of more than 99% was achieved when the samples were exposed to five laser shocks produced with laser pulses with energy more than 300 mJ (fig.5b). Cleaning under water resulted in higher efficiency because of the following reasons, 1) Greater magnitude of shock pressure due to confinement of plasma in water, 2) reduced adhesion force between the substrate
and the particulates by an order and 3) practically no redeposition when the samples were held vertically. The generation of cavitation bubbles in the liquid due to the focused laser beam also aids the cleaning process. Optical and electron microscopy of the cleaned surface revealed no damage on it.

Conclusion

Laser assisted surface cleaning has been implemented successfully in decontaminating metallic and dielectric contaminated substrates. This method is being used routinely in the AFFF fuel fabrication line for decontamination of PFBR fuel pins. Till date more than forty thousand fuel pins have been decontaminated using this technique. Other than removing loose contamination, lasers can play a dominant role in the decommissioning of aged nuclear reactors by ablating contaminated concretes and other metallic components. However, to exploit the full potential that lasers offers in surface cleaning a well directed research and development work is prerequisite.

References