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# $CO_2$ laser assisted removal of $UO_2$ and $ThO_2$ particulates from metal surface

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ABSTRACT

Pulsed laser assisted removal of uranium dioxide and thorium dioxide particulates from stainless steel surface have been studied using a TEA $CO_2$ laser. Decontamination efficiency is measured as a function of laser fluence and number of pulses. Threshold fluence for the removal of $UO_2$ particulates has been found to be lower than that required for the removal $ThO_2$ particulates. Usage of a ZnSe substrate, that is transparent to the laser wavelength used here, enabled us to decouple the cleaning effect arising out of absorption in the particulates from that in the substrate and has contributed towards understanding the mechanism responsible for cleaning. The experimental observations are also corroborated by simple theoretical calculations.



## 1. Introduction

Laser assisted removal of particulate contaminants from metallic surfaces is an area of current research interest and has a strong bearing in various fields, e.g., semiconductor industry and conservation of ancient artworks, where surface contamination poses a major problem. Generally, pulsed Nd-YAG [1–3], excimer [4–6] and TEA $CO_2$ [7,8] lasers have been used as the cleaning tool for such studies. In nuclear industry too, the use of lasers to decontaminate radioactive metal surfaces has generated considerable interest as it is a dry and non-contact process producing very little secondary waste in addition to reducing the possibility of exposure to the personnel. Although there exist several reports of removal of radioactive oxide layers from metallic surfaces [9–11], the literature on laser assisted surface cleaning of radioactive particulate contaminants is rather scanty. Removal of such contamination from the external surface of the fuel tubes is an important step in nuclear fuel fabrication from the radiological safety point of view. Moreover, the particulates need to be removed without any damage to the thin walled fuel tube and release of any air-borne activity. Roberts et al. [12] reported removal of uranyl nitrate hexa-hydrate micro-crystals and $UO_2$ kernels (diameter 500 μm), typical feed material for pebble bed modular reactor, off the metal surface. However, most of the power reactors world wide utilizes $UO_2$ or a mixture of $UO_2$ and $PuO_2$ powder as the feed material. For the proposed Indian advanced heavy water reactor (AHWR), $UO_2$, $PuO_2$ and $ThO_2$ powders will serve as the feed material. During pellet loading operation, the surface of the fuel element gets contaminated with these powdery particulates. Particle sizes in these powders lie within several micrometer and, therefore, they adhere strongly to the clad surface by the short range attractive forces [13]. For the removal of these particulates from the clad surface, this strong adhesive force must be overcome. It is now well known that exposure of the contaminated surface to a short duration, high energy laser pulse can result in the generation of a thermal stress that, in turn, leads to the expulsion of the contaminant particulates [13].

In this paper we report the results of our study on $CO_2$ laser cleaning of stainless steel (SS) substrates contaminated with $UO_2$ and $ThO_2$ powders. Decontamination efficiency (DE), defined as the percentage of initial activity removed by laser irradiation, has been measured as a function of laser fluence and the number of pulses. Uranium and thorium being alpha emitters, the activity of the samples was measured using a ZnS(Ag) scintillation detector before and after laser irradiation. Threshold fluence for cleaning of $UO_2$ contamination was found to be lower as compared to that for $ThO_2$ contamination. This allowed the selective removal of $UO_2$ from a mixture of $UO_2$ and $ThO_2$ particulates on SS substrate for a fluence of 0.75 J/cm$^2$ of the incident laser beam.

## 2. Experimental

An indigenously made helium free TEA $CO_2$ laser [14] operating at a wavelength of 10.6 μm and capable of delivering a maxi-

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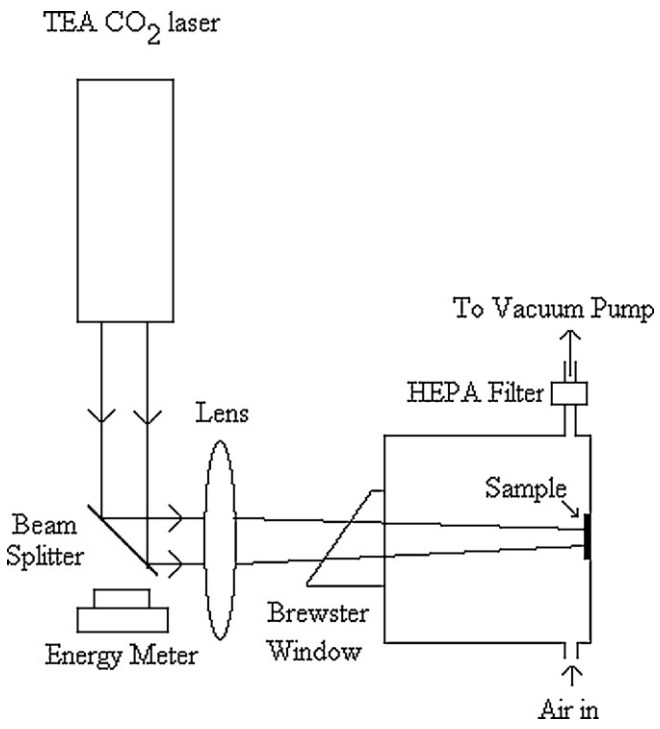


**Fig. 1.** Schematic of the experimental setup.

mum energy of 1.2 J over a pulse of duration 500 ns (FWHM) was used in these experiments. The laser emits a multimode beam of cross-section 1.5 cm × 1.2 cm. Energy of each pulse was measured by directing a small fraction of the beam to a pyro-electric joule meter (Gentec ED-320) with the help of an appropriate beam splitter. Schematic diagram of the experimental setup is shown in Fig. 1. Two ends of a cylindrical Perspex tube of length 15 cm and internal diameter 3.8 cm were vacuum sealed with the sample holder on one side and a ZnSe Brewster window on the other. Usage of an appropriately located ZnSe lens of focal length 30 cm allowed us to achieve the required laser fluence on the sample plane. The area of the contamination was maintained less than the beam size for all fluence values thus avoiding the necessity of scanning the sample across the incident beam. A rotary vacuum pump (Hindhivac-ED 21), connected to the chamber through a HEPA filter (10 cm × 10 cm × 8 cm) was continuously kept on during the experiment to avoid any air-borne activity in the working area. The samples were prepared by simulating $UO_2$/$ThO_2$ contamination on optically polished SS 316 (C: 0.35%, Cr: 18%, Ni: 12%, Mo: 2%, Si: 1%, Mn: 2%, rest Fe) and ZnSe discs of diameter 2.5 cm and thickness 0.2 cm. The contamination, in the form of powder taken along with a small quantity of isopropyl alcohol, was smeared on the surface of the substrate within a diameter of 8 mm. Usage of alcohol allowed transfer of sufficient activity minimizing, thereby, the error in counting. The average size of the $ThO_2$ particulates is 0.6–0.8 μm and the specific surface area of $ThO_2$ powder is 1.5 $m^2$/g with a bulk density of 0.7 g/$cm^3$. The average size of the $UO_2$ particles on the other hand is 2–3 μm with a specific surface area measuring 2.5–3 $m^2$/g and bulk density of 1.5–2 g/$cm^3$. Optical microscopy of the sample surface, however, revealed the presence of a range of particulate sizes due to the formation of agglomerates. For $UO_2$, the size of the agglomerates varied between 5 μm and 20 μm with a majority of them lying within 7–12 μm while for $ThO_2$ the agglomerate size varied from few micrometers to ~45 μm with an average size lying within 12–18 μm. The alpha activity of the samples was monitored before and after laser irradiation using a ZnS(Ag) scintillation detector (PLA-ALSCIN 991D). The samples were so chosen that the variation in their alpha activity lied within eight percent. The contamination so deposited is loosely bound to the substrate and represents the clad contamination of the fuel element following the pellet loading operation. The laser was operated in single shot mode during the experiment.

## 3. Results and discussion

In the first set of experiments, decontamination efficiency was studied as a function of number of laser pulses for samples with both types of contaminants for three different values of laser fluence, viz., 0.3 J/$cm^2$, 1 J/$cm^2$ and 2 J/$cm^2$ and the dependence is as shown in the traces of Fig. 2. For this study, always a fresh sample was used for each value of the fluence. Decontamination efficiency is seen to rise with increasing number of pulses until it saturates. Initial rise in DE with increasing number of pulses points to the fact that a fraction of the expelled particulates get re-deposited on the surface after each exposure. Re-deposition, as suggested by Kudryashov et al. [15,16], can be reduced either by using vacuum or a gas jet. It is to be noted here that a dynamic vacuum of ~100 mbar was maintained in the chamber during the course of experiments. The saturation behavior indicates that the cleaning force generated due to laser irradiation at this value of fluence is less than the adhesion force of the residual contaminants with the substrate and hence is unable to dislodge them. It is also seen from Fig. 2 that there is a marked difference in DE obtained for $UO_2$ and $ThO_2$ particulates at lower fluence levels. However with increasing fluence, the difference reduces and becomes vanishingly small for a fluence of ~2 J/$cm^2$.

Towards understanding this behavior, we attempted to decouple the effect of cleaning due to absorption of energy by the substrate from that by the particulates. To this end, we performed the next experiment by simulating the same contamination but now on ZnSe substrate that is transparent to 10.6 μm wavelength. Fig. 3 shows the dependence of DE for $UO_2$ and $ThO_2$ particulates on number of pulses for a fluence of 1 J/$cm^2$ in this case. It is seen that irradiation does not result in any significant cleaning of $ThO_2$ particulates, while considerable cleaning of $UO_2$ particulates is obtained. It is therefore apparent that cleaning arising due to absorption in the particulates alone is significantly lower in case of $ThO_2$ as against $UO_2$. This inference is further corroborated by direct absorption measurements in the particulates. The absorption was estimated by comparing the transmission of the laser beam through ZnSe plate with and without contamination. It was found that while $UO_2$ particulates exhibited an absorption of ~90%, $ThO_2$ particulates showed only ~20% absorption. In light of these findings, a qualitative explanation can be offered to the observations made by us with regard to the cleaning of $UO_2$ and $ThO_2$ contamination on metal surfaces.

The higher DE obtained in the cleaning of $UO_2$ contamination for all fluence values is primarily due to the large absorption of incident radiation by the particulates. That the substrate does not play an important role in the removal of $UO_2$ particulates is also evident from the observation of same cleaning efficiency for both ZnSe and SS substrates (Fig. 4a). As the absorption of incident radiation by $ThO_2$ particulates is poor and SS substrate too has negligible absorption at the wavelength of the incident light, efficiency of removal is poor at lower fluence. With increasing fluence, the increased cleaning force as a result of the correspondingly increased absorption, both in substrate and the particulates, results in higher DE. The contribution of the substrate in cleaning $ThO_2$ particulates is evident from Fig. 4b. Average particle size of $UO_2$ is ~3 μm, where as that of $ThO_2$ is less than 1 μm. Smaller particle size along with the platelet morphology of $ThO_2$ particulates [17] results in a much stronger adhesion with the substrate in comparison to $UO_2$ partic-