Optics and Laser Technology 106 (2018) 286–293

Contents lists available at ScienceDirect

Optics and Laser Technology

journal homepage: www.elsevier.com/locate/optlastec

Full length article

Particulate size and shape effects in laser cleaning of heavy metal oxide loose contamination off clad surface

Aniruddha Kumar [a,*], D.J. Biswas [b]

[a] Advanced Fuel Fabrication Facility, Bhabha Atomic Research Centre, Tarapur 401 504, India
[b] Laser & Plasma Technology Division, Bhabha Atomic Research Centre, Mumbai 400 085, India

A R T I C L E I N F O

Article history:
Received 26 November 2017
Received in revised form 18 March 2018
Accepted 25 April 2018

Keywords:
Laser cleaning
Morphology
ThO_2
UO_2
Wavelength

A B S T R A C T

This work explores laser assisted removal of uranium-di-oxide and thorium-di-oxide particulates from zircaloy substrate. An electro-optically Q switched Nd-YAG laser operating on three wavelengths of 1064 nm, 532 nm, 355 nm was used as the cleaning tool to compare the efficiency of the decontamination process. Laser assisted surface cleaning that offers the dual advantage of remote operation and minimizing the generation of secondary waste has therefore emerged as the most attractive technique with regard to decontamination of radioactive surface. In case of loose contamination wherein the contaminant particulates are often attached to the substrate surface by short range van-der-wall's force, the absorption of energy from the incident radiation field by both particulates and substrate can contribute towards generation of cleaning force leading to expulsion of the particulates. It was found that laser parameters like its wavelength, fluence, number of exposures and particulate shape/size play an important role in the cleaning process. A single laser exposure with a fluence of ~0.3–0.5 J/cm^2 was found to be enough to obtain decontamination efficiency of ~70–80% while removing the contaminants from zircaloy substrate. Subsequent exposure of laser pulses to the contaminants further increases the efficiency of cleaning. Decontamination efficiency was found to be always higher for uranium-di-oxide particulates than virgin thorium-di-oxide particulates under identical cleaning conditions. However, decontamination efficiency of milled thorium-di-oxide particulates was found to be marginally higher than uranium-di-oxide particulates.

1. Introduction

India is pursuing a three-stage nuclear power programme for effective utilization of uranium and thorium reserves to fulfill the ever-growing need of energy [1]. This three-stage program is based on a closed fuel cycle, where 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 second stage comprises of Fast Breeder Reactors (FBR's). In the second stage, by reprocessing the spent nuclear fuel from PHWR's and using the recovered plutonium in fast breeder reactors, the non-fissile depleted uranium and thorium can breed additional fissile material plutonium and uranium-233 respectively. The third stage is based on the operation of thorium and uranium-233 fueled nuclear reactors. An Advanced Heavy Water Reactor (AHWR) will be effected in the third stage. AHWR will utilize (Th, U-233) O_2 as the fuel material and zircaloy 2 as the clad material [2]. The fuel for AHWR needs to be fabricated inside heavily shielded glove boxes or inside a hot cell owing to very high gamma dose rate normally associated with it. Operations involving fuel fabrication thus need to be mechanized with minimum manual intervention [3]. Following the loading of the fuel tubes with MOX (Mixed Oxide) fuel pellets, the outer clad surface gets contaminated with loosely attached radio-toxic particulates necessitating their cleaning before they can be removed from the glove box/hot cell for further processing and assembly. We envisage the laser assisted surface decontamination to play an important role as the laser beam can be readily transported inside the glove box or hot cell offering the prospect of cleaning of AHWR fuel elements in a remote and dry manner. Laser assisted removal of particulate contaminants is an area of current research as it provides a fast, efficient and non-contact process of cleaning metallic as well as dielectric surfaces. These advantages have resulted in employing laser-based surface cleaning process in many areas of engineering and science, e.g., nuclear industry [4–9], semiconductor industry [10–12], restoration of art works [13–15] and manufacturing industries [16,17]. Particulates with size in the range of micron are adhered to the substrate by short range attractive forces like Van der Waal's force

* Corresponding author.
E-mail address: aniruddhakumar@barctara.gov.in (A. Kumar).

https://doi.org/10.1016/j.optlastec.2018.04.027
0030-3992/

that can exist between both polar and non-polar substances [18]. The strength of adhesion 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 giving rise to the generation of a thermo-elastic force. The particulates can get dislodged from the substrate when the generated force exceeds the adhesion force [19]. The wavelength of the laser as well as the physical properties of the particulate and substrate like thermal conductivity, specific heat density, thermal expansion coefficient etc. is known to have a strong bearing on the generated cleaning force. We present here results of our experiment on the laser assisted removal of ThO_2 and UO_2 particulates off metal substrate aimed at gaining deeper insight into the dependence of wavelength and fluence of the exposure as well as the particulate morphology on the cleaning process.

2. Materials and methods

2.1. Equipment

An electro-optically Q switched Nd-YAG laser (EKSPLA-NL313) capable of delivering a maximum of 1.6 J, 800 mJ and 500 mJ respectively on 1064 nm, 532 nm and 355 nm over a pulse of duration 6–8 ns was used as the cleaning source for this experiment. The laser emits a polarized beam of spatial cross section ~1 cm^2 with top hat intensity profile (Fig. 1). The irradiance used in our experiment was $1.2 - 6.2 \times 10^7$ W/cm^2.

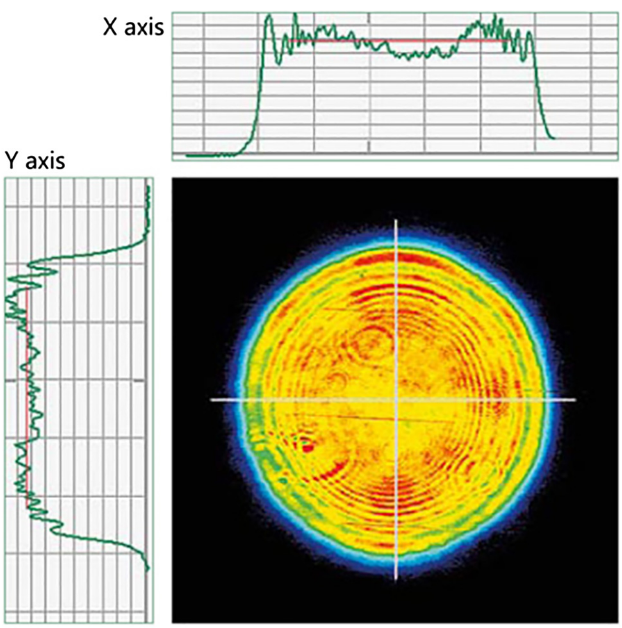

Fig. 1. Top hat intensity profile of the laser beam as recorded by a beam analyzer. The near flat top spatial character of the laser beam is clearly evident from the X and Y scans as shown in the figure.

2.2. Preparation of samples

Three different types of powders were used as the test contaminant for this study viz., (1) UO_2 powder with irregular morphology, (2) virgin ThO_2 powder with platelet morphology and (3) milled ThO_2 powder with irregular morphology and are depicted in the traces 'a–c' respectively of Fig. 2. The contamination was simulated on the surface of zircaloy substrate of dimension 15 mm × 15 mm × 0.4 mm smearing the contaminant powder taken along with a small quantity of isopropyl alcohol over a diameter of 7–8 mm (Fig. 3a). The average size of UO_2 and ThO_2 particulates lied in the range of 0.6–1 μm and 0.3–0.5 μm respectively. Scanning electron microscopy (Model: FEI Quanta 200SEM) of the sample surface revealed the presence of a range of particulate with varying sizes basically due to the formation of agglomerates. For UO_2 the size of agglomerate varied between 6 μm and 20 μm while for ThO_2 the agglomerate size lied within few μm to 12 μm.

2.3. Experimental set up

The schematic of the experimental set up is shown in Fig. 4. The samples so prepared were then placed inside a Perspex-chamber, one end of which was sealed with the sample holder and the other end with a LiF Brewster window that allowed complete transmission of all the wavelengths used for this investigation. The laser beam always intercepted the entire contaminated area of the sample (Fig. 3b) thus avoiding the necessity of any scanning. The Perspex chamber was placed ~100 mm away from the laser aperture.

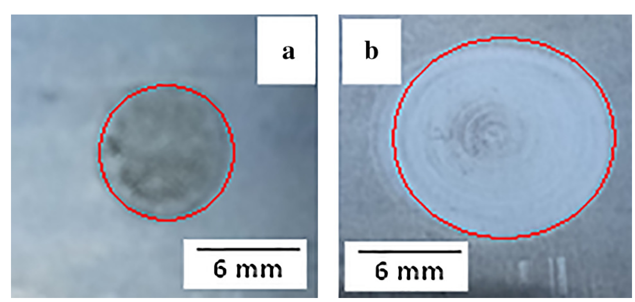

Fig. 3. (a) Contaminated area on the zircaloy sample and (b) the same area marked in trace "a" after laser exposure is recorded here. This clearly reveals complete interception of the contamination by the laser beam.

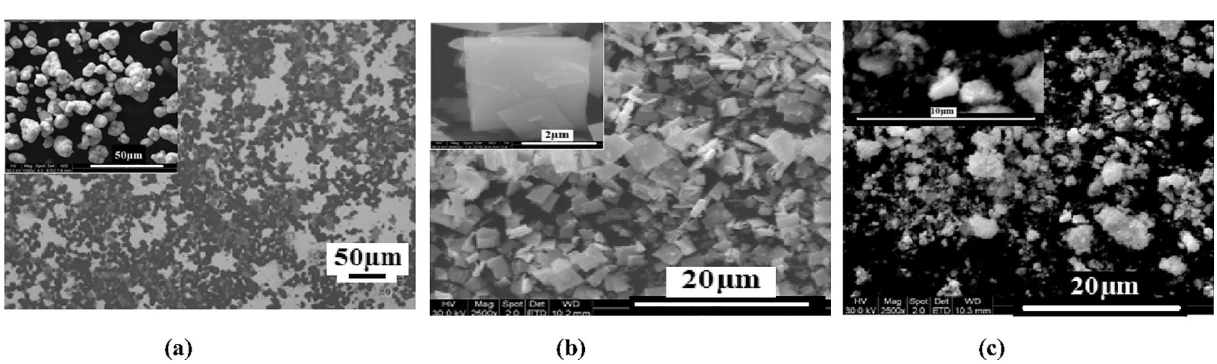

Fig. 2. SEM images of the contaminants, (a) UO_2 powder, (b) virgin ThO_2 powder, and (c) milled ThO_2 powder. Magnified view is recorded as inset to these images.