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Laser assisted removal of fixed radioactive contamination from metallic substrate

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HIGHLIGHTS

- Fixed contamination on metallic substrate was removed.
- Contamination was due to presence of caesium and cobalt isotopes.
- A pulsed fiber laser was used as the removing tool.
- Laser ablation was the mechanism of removal.

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ABSTRACT

Metallic waste contaminated with radioactive materials accounts for a large proportion of the nuclear wastes generated in nuclear facilities. Storage of these highly contaminated wastes is known to attract high cost. Reduction of activity of these wastes can significantly bring down the cost of their storage. Further, for recycling of the precious metal, it's mandatory that it's surface contamination is first brought down to the accepted level. We report here a practical method of significant reduction of volumes of contaminated wastes to be stored by way of employing coherent radiation emanating from a laser to ablate fixed radioactive contamination alone from the surface of metallic wastes. A single mode pulsed fiber laser was used to remove fixed contamination from stainless steel substrate by ablation. Optimisation of the removal process was effected with respect to the laser power, repetition rate, laser beam scanning speed and number of passes. Ablation depths ranging from few microns to few hundreds of microns could be readily achieved as per the requirement simply by suitably varying the above processing parameters. Samples simulated with U-232 contamination as well as samples from the pipe line of a Boiling Water Reactor (BWR) were found to be decontaminated containing only marginal residual activity using this technique.

1. Introduction

Metallic wastes contaminated with radioactive material account for a significant proportion of nuclear wastes generated in major nuclear facilities like nuclear power plants, fuel fabrication and re-processing plants. These contaminated materials are normally disposed by reducing the residual activity by decontamination (Valencia, 2012). The main aim of decontamination is to reduce the contamination of the components to such levels that they can be disposed of at a lower, and therefore more economical, waste treatment and disposal category or indeed disposed of as waste exempt from regulatory concern. The contaminants here can be both loose, often attached to the substrate by weak van der Waal's force (Nilaya et al., 2008) or fixed wherein it is either chemically bonded to or trapped inside pits/scratches of the substrate (Dabrowski, 2001). While loosely attached radioactive particulates can be efficiently dislodged from metallic surfaces by both ultrasonic (Kumar et al., 2014) and laser (Kumar et al., 2013) sources, the removal of fixed contaminants is more involved as it involves breaking of chemical bonds before the contaminated layer can be physically removed from the subject. A typical example of fixed contamination is the one present both on the reactor

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core components and on the primary structural materials in primary cooling circuit. In the primary circuit of water cooled nuclear reactors there are three main sources of radioactive contamination 1) fission products and actinides released from a leaky or defective fuel element, 2) activation of feed water and its impurities and 3) activation of various corrosion products (Hirschberg et al., 1999). In these nuclear reactors, the major part of radioactive contamination is formed when corrosion products are released from primary structural materials, transported to in-core regions and turns radioactive by activation. A part of these corrosion products gets deposited on the oxidized surface of re-circulation pipes and other core components. The primary radio nuclides which contribute to the radiation field in primary side coolant are Co^{58}, Co^{60}, Cs^{137}, Cr^{51}, Fe^{59}, Mn^{54} (Bodansky, 2004; Varga et al., 2001). Selective removal of these contaminants from the structural components inevitably reduces significantly the volume of the contaminated metallic waste. A number of cleaning methods (Technical Reports Series No 401, 2001) mostly based on wet techniques, e.g. chemical cleaning, electro-chemical cleaning and wet abrasive blasting methods are presently employed in nuclear industry to remove the fixed contamination from metallic components. However, handling of toxic and corrosive chemical reagents along with generation of relatively high volumes of secondary liquid waste is a concern in all the above mentioned techniques. In some cases it is also difficult to control the amount of material substrate removed in these processes (https://www.oecd-nea.org/rwm/reports/1999/decontec.pdf). To this end laser has emerged as the most versatile tool over the years as it has the ability to effect localized heating of a preselected region leaving the virgin substrate underneath practically untouched. Besides being a dry and non-contact process, the laser assisted cleaning has the inherent advantage of remote operation as the laser beam can be easily transported through transmittive optics or by optical fiber inside the glove box or the hot cell without any manual interference (Nilaya et al., 2006). The ability of lasers to deposit large amount of energy in pulses of short duration on the surface may result in generation of thermo-elastic stress necessary to remove loose contamination or extreme localized heating leading to ablation of a thin layer of fixed contamination from the substrate surface (Zhou et al., 2002). Laser ablation is the process of removal of material from a substrate by absorption of laser energy. The efficiency of this process is described by the ablation rate which can be defined as the volume or mass or thickness of material removed per pulse during irradiation with a laser pulse. The onset of ablation in a material occurs only when the fluence is above a certain threshold value. The threshold value of fluence depends upon the laser wavelength, pulse duration and material properties. Once the fluence is above threshold, volume of material removed per pulse typically increases with increase in fluence values. The material from the substrate is removed by different mechanisms depending on the laser fluence value. At lower fluence values, the mechanism is evaporation whereas at higher fluence values the mechanism is phase explosion. Laser's temporal pulse length has a significant effect on the dynamics of laser ablation. Threshold value of fluence for ablation of a material reduces at shorter pulse duration. Thermal damage of the substrate material increases with increase in pulse duration. As the volume of the material removed per pulse is significantly low, it is necessary to use a laser operating at a high repetition rate to obtain significant material removal from the substrate. Material from a large area is removed by scanning the focussed beam over the surface using motorised galvo- scanners. For a fixed beam scanning speed and repetition rate, increase in laser power increases pulse energy, thereby increasing the ablation depth per pass. Whereas operating the laser at fixed power and repetition rate, increase or decrease in scan speed decreases or increases the number of interacting pulses per unit length of the substrate respectively, thereby decreasing or increasing the ablation depth per pass. Thus, laser ablation is a process which can be used to decontaminate metal surfaces where the contamination is linked with the substrate by a chemical bond or the contaminant is trapped inside the pits/scratches on the substrate.

We report here the results of our work on removal of fixed contamination from the internal surface of a stainless steel pipe used for coolant level indication in a Boiling Water Reactor. A single mode pulsed fiber laser was used as the coherent source for this investigation. Several precursory experiments were carried out with simulated contamination to optimise the laser parameters before moving on to the actual samples collected from the reactor. For the precursory studies, contamination was simulated by electro-depositing trace amount of U-232 on stainless steel substrate. A judicious combination of the laser and beam scanning parameters allowed achieving ablation depth that could be varied from few micrometers to few hundreds of micrometers. Following laser ablation, the activity of the sample was monitored by gross alpha counting using a scintillation detector. After optimisation of the parameters, stainless steel samples from the contaminated coolant pipe line were decontaminated using the laser. The major activity of the samples was found to be due to the presence of Cs^{137} and Co^{60} nuclides. Caesium is produced by nuclear fission in the reactor fuel and usually contained within the cladding of the nuclear fuel tubes. If the cladding is damaged then caesium can get released from the defective fuel element and reacts with water to form caesium hydroxide. Caesium hydroxide is highly reactive and reacts with stainless steel to form isolated caesium cations. These cations get incorporated in the oxide layer by ion exchange formed over the steel substrate (Ajlouni et al., 2010). Significant quantity of cobalt is found in the components which are subjected to mechanical wear and are usually made from hard facing alloys e.g. Hastelloys (Prince, 2012). Cobalt in trace quantity is also present in different structural materials e.g. various types of stainless steel, zircaloy and inconel. The origin of Co-60 is through n-γ reaction with Co-59, natural constituent of cobalt. Co-60 is γ emitter, and activity of these samples was measured by a NaI detector. The ablation depth on the samples was monitored by a surface profilometer.

2. Experimental

A single mode pulsed fiber laser (pulse duration 30 nsec FWHM; rep. rate: 1 Hz–1 MHz) capable of delivering up to 50 W average power and operating at 1060 nm wave length was used as the decontamination tool for this study. The laser beam could be focussed on the job by using an F- theta lens of focal length 156 mm to a diameter of ~50 μm. The surface of the job was placed at the focal plane and a set of motorised galvos were made use of to scan the required surface area by the focussed laser beam. The scanning speed could be varied between few mm/s to few hundreds of mm/s. The samples (diameter: 30 mm, thickness: 5 mm) in the form of solid disc for the precursory experiments were prepared from SS 304 rod by machining. The central portion of these samples (around 15 mm in diameter) was contaminated with U-232 by electro- deposition technique. The initial activity of the samples was measured by gross alpha counting using a ZnS –Ag scintillation detector. The samples simulate the condition of fixed contamination in oxide layers of primary cooling loops of a reactor system. The samples for the second set of experiments were prepared from a primary coolant pipe line (SS 304, outer diameter: 28 mm, thickness: 3 mm) of a Boiling Water Reactor. Firstly, the pipe was cut axially into two equal halves and then each half was further cut to small pieces of size admeasuring approximately 25 mm × 25 mm. The curved area on the inner surface of the pipe was expected to be contaminated with activated corrosion products.