Radiation Physics and Chemistry 95 (2014) 188–190

Contents lists available at ScienceDirect

Radiation Physics and Chemistry

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

Laser decontamination of the radioactive lightning rods

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HIGHLIGHTS

- The process generates minimal additional secondary waste.
- The effectiveness of this technique may allow certain materials to be recycled reducing radioactive waste volumes.
- The process allows reuse of decontaminated metals.

ARTICLE INFO

Article history:
Received 28 September 2012
Accepted 22 March 2013
Available online 30 March 2013

Keywords:
Radioactive lightning rods
Laser surface decontamination
Laser ablation
Radioactive waste

ABSTRACT

Between 1970 and 1980 Brazil experienced a significant market for radioactive lightning rods (RLR). The device consists of an air terminal with one or more sources of americium-241 attached to it. The sources were used to ionize the air around them and to increase the attraction of atmospheric discharges. Because of their ineffectiveness, the nuclear regulatory authority in Brazil suspended the license for manufacturing, commerce and installation of RLR in 1989, and determined that the replaced RLR were to be collected to a centralized radioactive waste management facility for treatment. The first step for RLR treatment is to remove the radioactive sources. Though they can be easily removed, some contaminations are found all over the remaining metal scrap that must decontaminated for release, otherwise it must be treated as radioactive waste. Decontamination using various chemicals has proven to be inefficient and generates large amounts of secondary wastes. This work shows the preliminary results of the decontamination of ^{241}Am-contaminated metal scrap generated in the treatment of radioactive lightning rods applying laser ablation. A Nd:YAG nanoseconds laser was used with 300 mJ energy leaving only a small amount of secondary waste to be treated.

1. Introduction

Several hundred radioactive lightning rods (RLR) manufactured with radium sources were imported to Brazil until early 1970 when cheap ^{241}Am sources became available and the manufacture of RLR became economically feasible. It has been estimated that around 75,000 RLR were manufactured and installed over the roofs of factories, commercial buildings, high storied residential buildings, schools, and other structures while the license for use was in place, between 1970 and 1989.

The proponents of this technology claimed that the ionization of the air around the rods reduced the resistivity of air, creating a stream of ascending charges that improved the attraction power of the rod and widened the protected area. It was only in mid-80s that the efficacy of the radioactive early streamer emission air terminals, as they are technically classified, were definitively dismissed by experts (Baigalmaa et al., 2009), although strong evidence contrary to the effect of ionizing radiation on the incidence of lightning bolts had been presented at last two decades earlier (Gillespie, 1965).

Following the example of other countries, in 1989 the nuclear regulatory authority in Brazil suspended the license for manufacturing, commerce and installation of RLR and determined that the replaced RLR were to be collected to a centralized radioactive waste management facility (CNEN—Comissão Nacional de Energia Nuclear, 1989).

The twenty thousand RLR which have been collected until now and have been treated as radioactive waste showed at least a detectable level of surface contamination, after the attached sources had been removed. A significant fraction of the RLR was actually contaminated well above clearance levels and was refractory to simple decontamination methods. Although the sources were originally manufactured as sealed sources, they were susceptible to some degree of deterioration after many years of exposure to the elements, causing part of the radioactive material to disperse.

Brazilian-made RLR were manufactured with one to ten ^{241}Am sources, depending on the expected protection area required by

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http://dx.doi.org/10.1016/j.radphyschem.2013.03.043

the customer, each source with average activity around 7 MBq (Minematsu et al., 2009). The sources were supplied by the former Amersham plc® and were made with a layer of americium oxide deposited over a stainless steel ribbon and covered with a thin layer of a gold alloy that encapsulated the radioactive material to avoid its dispersion. Sections of the ribbon were cut and fastened to strips of stainless steel which were in turn riveted to the plates of the air terminals. Two of such sources are visible in the upper surface of the bottom plate of the RLR shown in Fig. 1. Note the rivets in the top plate which are evidence of other sources attached to the RLR shown.

The encapsulating layer of the sources had to be thin enough to allow alpha particles to be emitted without absorption and had to be made of noble metal to resist corrosion in the outdoor environment. Whether by negligent manufacture and installation or by wind erosion; however, the surface of the sources is usually dotted with microscopic punctures and scratches (Marumo et al., 2008) through which a bit of the source material escapes and contaminates the rods and the surrounding structures.

Although the contamination levels found pose no immediate risk to public health, the release of radioactively contaminated materials is regulated and the metal scrap must be previously decontaminated for release or otherwise must be treated as radioactive waste. The decision to hold back or to release rests on a comparison between the costs of decontamination and the costs of treatment as radioactive waste.

The second alternative above for managing the metal scrap from the treatment of the RLR is somewhat expensive, considering the number of lightning rods to be treated. Thus, the application of a decontamination technology that is inexpensive and that allows achieving the required decontamination factors is a key point in the management of this radioactive waste. For more than fifty percent of all RLR collected until now there is a bonus in the decontamination option, which is the possibility of recycling one to two kilograms of copper alloy of each RLR. Other materials are stainless steel and tin plated carbon steel, which are less attractive for recycling, yet still indicated for decontamination and release.

Decontamination tests using a variety of chemicals were carried out previously (Dellamano et al., 2009; Fonseca and Dellamano, 2011) and the results, yet satisfactory from the point of view of the decontamination factors achieved, showed that a more efficient method should be sought to avoid the large amounts of decontamination solutions left as secondary waste.

In this work we aim to present the results of tests with laser ablation as a method for decontamination of the ^{241}Am-contaminated metal scrap generated in the treatment of radioactive lightning rods.

2. Experimental setup

The laser used in this work was a nanosecond Nd:YAG (Quantell Brilliant), operating in the fundamental mode, 1064 nm, with a pulse duration of 5 ns and 300 mJ per pulse. No optical element was used to focus the laser beam in order to keep a long confocal parameter, thus avoiding that sample reliefs interfered in the spot size, which was 3 mm in diameter. This laser was selected after indication of the literature (Veiko et al., 2011). Due to the almost disk-like symmetry of the plates the laser beam was maneuvered with two dielectric plane mirrors (Layertec High Power) designed for 1064 nm, mounted in kinematic supports that allowed the beam to scan entirely the surface of each face of the sample as shown in Fig. 2. One of the mirrors was connected to a lever driven by a computer-controlled step motor and the plate was placed inside a specially built acrylic sealed box and made to rotate around its axis by another step motor.

The laser beam emerging from the laser head (left side of Fig. 2), was maneuvered with the mirror scanning system and injected through a glass window into the sealed box, where the sample was processed. Pressure inside the box was maintained below atmospheric pressure with a general purpose laboratory vacuum pump to prevent radioactive material from leaking. A high efficiency particulate air (HEPA) filter was fit at the box exhaust port and connected to the vacuum line thus avoiding contamination of the pump.

The 59 keV gamma emission of Am-241 from the RLR plates was measured before and after laser ablation with a Hyperpure Germanium Detector (HPGe) from Canberra, model GX2518, and electronic setup composed of high voltage source, amplifier and multiport multichannel analyzer. The counting geometry was detector and plates coaxially mounted at 10 cm.

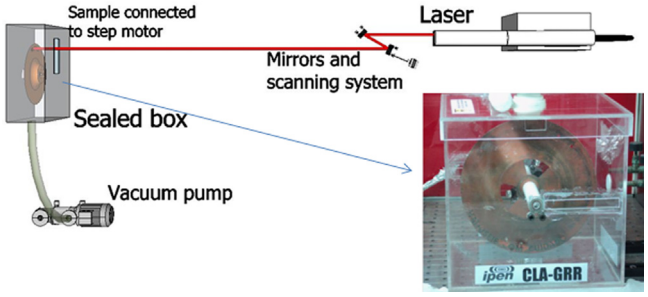

Fig. 2. Decontamination system setup and RLR decontamination box.

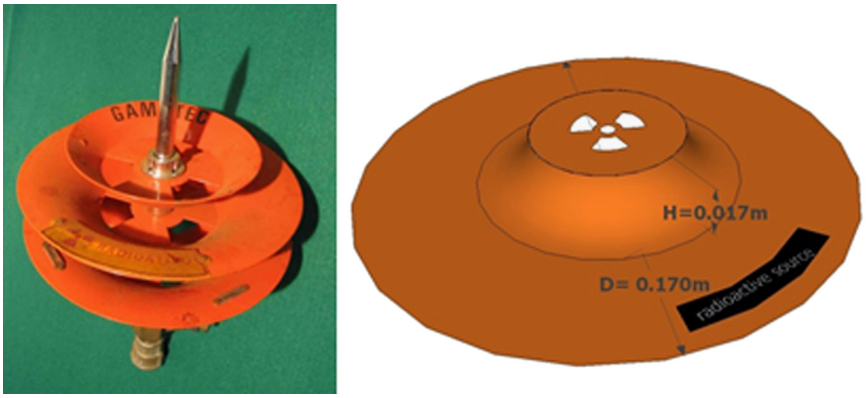

Fig. 1. RLR plate (left) and schematic RLR (right).

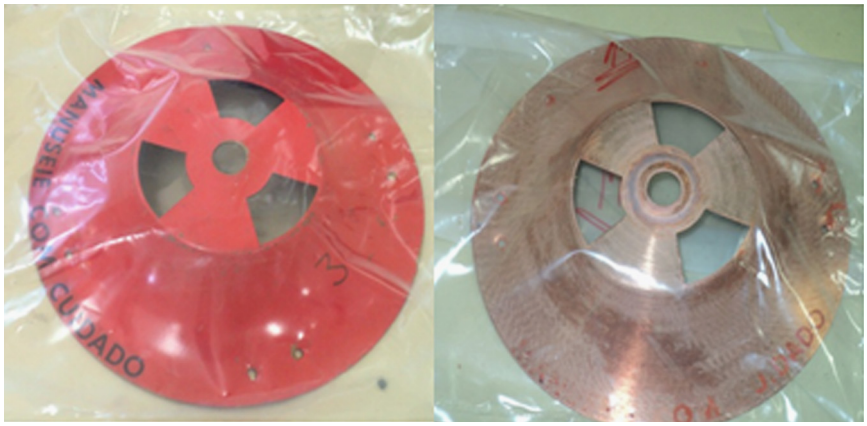

Fig. 3. A RLR plate, before (left) and after (right) laser cleaning and decontamination.

Table 1
Decontamination factors obtained with laser cleaning.

Plate	Surface	Before cleaning (cpm)	After cleaning (cpm)	DF[a] (%)
1	Upper	33771	3875	89
	Lower	8962	1662	81
6	Upper	13773	286	98
	Lower	10754	199	98
4	Upper	18218	902	95
	Lower	1858	162	91
2	Upper	8909	4723	47
	Lower	3207	2358	26
5	Upper	4036	566	86
	Lower	1077	212	80
8	Upper	123	3	97
	Lower	65	2	96
7	Upper	812	129	84
	Lower	209	28	87
3	Upper	10501	4006	62
	Lower	4144	1805	56

[a] DF=Decontamination Factor

3. Results and discussions

Fig. 3 shows one RLR painted copper plate before and after the laser cleaning process. The laser ablation stripped of the painting almost completely. The scanning of each surface lasted 25 min. The coating was removed as peeled back chunks or as particulate material with varying grain sizes that were ejected from the plate surface by the expansion of the plasma produced by the laser. These particles exhibited large initial velocities and settled onto all box walls. A fine aerosol was also formed by the condensation of vaporized paint and was retained mostly by the HEPA filter.

The particulate residues of each decontamination run were later removed from the box and processed by acid leaching to form a solution for activity determination.

Table 1 shows counting results for both sides of eight plates before and after laser ablation and the decontamination factors were calculated as the percent of the ^{241}Am activity removed. The higher initial contamination levels were always on the side of the plates where the americium sources were formerly riveted.

The scanning of smooth surfaces always resulted in very high decontamination factors but the area of the plates around the holes left by removing the rivets was usually a bit damaged and lower decontamination efficiencies were obtained in this area. Remnants of painting and crevices, fissures and cracks in the plates retained particles of americium oxide which resulted in the lower decontamination factors observed. This problem is tackled by adjusting the laser beam focus and other operational parameters as to optimize the process and achieve complete removal of the contamination.

The secondary wastes generated in this decontamination process were only the stripped paint and the HEPA filters that must be replaced after some time of operation. Both presented a very low mass and volume and can be treated as compactable radioactive waste.

4. Conclusion

Laser cleaning was found to be a powerful method for radioactive surface decontamination. The results showed that this process is effective and generates the lowest volume of secondary waste of all decontamination process previously evaluated for treatment of RLR scrap metal. Moreover, it allows the reuse or recycling of the decontaminated metal, thus reducing radioactive waste volumes and management costs.

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