

Surface Decontamination Using LASER Ablation Process – 12032

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ABSTRACT

A new decontamination method has been investigated and used during two demonstration stages by the Clean-Up Business Unit of AREVA. This new method is based on the use of a LASER beam to remove the contaminants present on a base metal surface. In this paper will be presented the type of LASER used during those tests but also information regarding the efficiency obtained on non-contaminated (simulated contamination) and contaminated samples (from the CEA and La Hague facilities). Regarding the contaminated samples, in the first case, the contamination was a quite thick oxide layer. In the second case, most of the contamination was trapped in dust and thin grease layer. Some information such as scanning electron microscopy (SEM), X-Ray scattering spectroscopy and decontamination factors (DF) will be provided in this paper.

INTRODUCTION

Dismantling and decommissioning (D&D) operations are usually complex and lengthy; they are an important step in the life cycle of nuclear facilities and require the use of different kinds of skills and technologies. The choice of the best one involves to take into account numerous parameters such as the economical (cost), physical (size and accessibility of cells to be decontaminated), radiological (type and level of contamination, wastes production, operators dosimetry) and organizational & human factor (OHF).

About surface decontamination, it is commonly admitted that 80% of the metal waste are only surface contaminated [1].

Because most of radioactive contaminants are usually incorporated into a thin oxide layer, if we focus on metal surfaces decontamination, the methods commonly used are dedicated to the removal of such oxide layer [2-5]. Common methods used to achieve this goal are blasting (sand, dry ice, metallic shots), disk sander, electrodecontamination, or chemical decontamination (CORD [3,6], EMMAC [7], gels [8,9], foams [8,10]...). Such techniques have been successfully used to decontaminate nuclear surfaces for many years. However, for some of them, efficiency is somehow limited (e.g, Dry Ice blasting) for others, the wastes production (liquid and/or solid) remains an important problem.

To answer to all these issues, the Clean-Up Business Unit of AREVA decided to work on the development of a new technology: the LASER Ablation. This technology, based on the interaction between a LASER beam and the surface of the base metal, can be defined as a "dry" technology. It means that no liquid waste is produced. In parallel, this method also allows to avoid any secondary waste production.

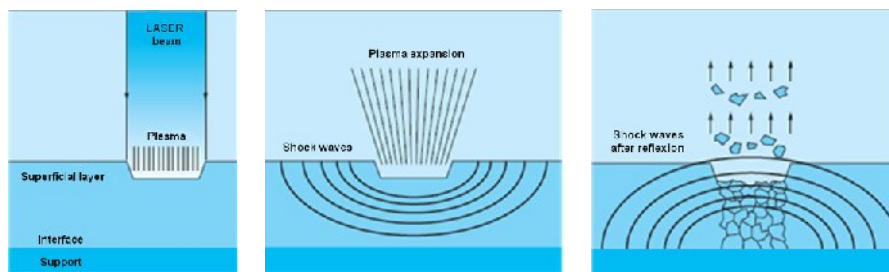
To deploy this technology, two ways can be used such as the manual one or an automatic one. In this case it allows to reduce occupational exposure to workers and to increase the productivity. Finally, it is possible to add on the system a nozzle, linked to a vacuum system to vacuum of dusts removed from the metal base surface during the process.

Since 2007, several tests have been performed, firstly in inactive and secondly, in active conditions, to demonstrate how this technology can be efficient to achieve the decontamination of materials even in presence of strongly fixed contamination. In all our tests, experiments were carried out using the Nd:YAG (neodymium-doped Yttrium Aluminium Garnet) LASER technology.

OVERVIEW OF LASER TECHNOLOGY

When a metallic surface is irradiating by a YAG LASER beam, the surface will absorb energy generated by the incoming photons [11]. If this absorption is efficient, we can assist at the conjugation of two main effects: thermal and mechanical (Scheme. 1).

Regarding the thermal effect, the quick increase of the surface temperature, due to the interaction between the LASER beam and the metallic surface, will lead to the plasma formation. After that, the plasma expansion will generate some mechanical waves which will propagate into the surface to crack and thus, causes the ejection of contaminated particles. That is the "mechanical effect". Until the surface is totally clean, the emitted LASER beam will be almost completely reflected. Thus, the process will stop.



Scheme 1. LASER ablation principle.

MATERIAL USED FOR EXPERIMENTS

LASER

As mentioned previously, the choice to use the YAG technology to carry out our evaluations has been made. Two different LASER units were used during our experiments:

- The first one was a LASER optically pumped with flash lamps and with a power of 36 watts.
- The second one was also optically pumped but with diodes; the power of this second equipment was 300 watts.

In both cases, the wavelength was centered on 1064 nm (near infrared domain) and a vacuum system has been added on or close to the LASER optic. This allows to trap the removed contamination and avoid any dissemination in the cell.

Samples for Inactive Conditions

Two different series of experiments were carried out in non-radioactive conditions. What we have used for the first one were stainless steel plates coated with rust, paints or grease. For the second series, metallic plates covered with radionuclide simulants¹ such as Cs_2CO_3 , SrCO_3 , RuO_2 , $\text{UO}_2(\text{NO}_3)_2$, $\text{Th}(\text{NO}_3)_4$ have been used.

Samples for Active Conditions

As we did previously, two series of experiments have been done in active conditions. The first one, on tank samples from CEA of Grenoble and the second one, on metallic pieces from AREVA La Hague. In the first case, samples were covered with a thick oxide layer (metallic oxides oxide formed under ambient pressure and temperature with presence of nitric acid over many years). In the second case, samples were covered essentially with a thin grease layer.

RESULTS

This part will present the results obtained for each series of experiments starting with the ones obtained in inactive conditions especially on rust.

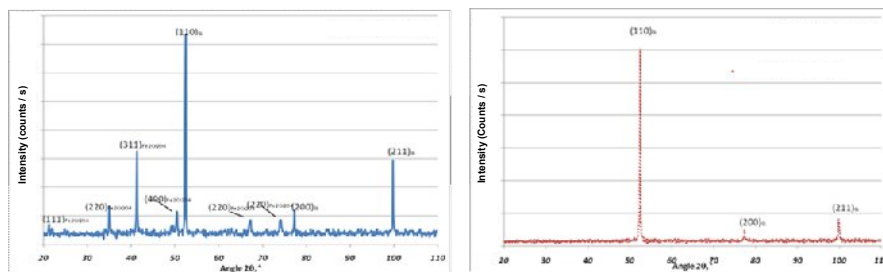
Results Obtained in Inactive Conditions

As it is shown on figure 1, LASER process seems to be really efficient to remove an iron oxide layer without inducing any structural modifications of the substrate.



Fig. 1. Ablation of iron oxide (Before treatment (Left), After treatment (Right)).

As we can see on graphic 1, the LASER beam does not induce modification into the metallic surface. The comparison of X-Ray spectrum (before (Left) and after treatment (Right)), shows that the only difference is the disappearance of signals related to the rust, especially the chromite FeCr_2O_4 (signals: 111, 200, 311, 400 and 220).



Graphic 1: X-Ray scattering analysis of iron oxide ablation on carbon steel.

¹ The salts were encrusted onto the surface by a dry friction.

On simulated contamination, the results of this technology appear to be also really promising. Indeed after adding various types of radionuclide salts onto the test surface, a LASER beam with an energy density of 1 J.cm^{-1} has been emitted.

The influence of the pass number has been investigated. The process efficiency was determined by using scanning electronic microscopy analysis. The results are shown below on figure 2.

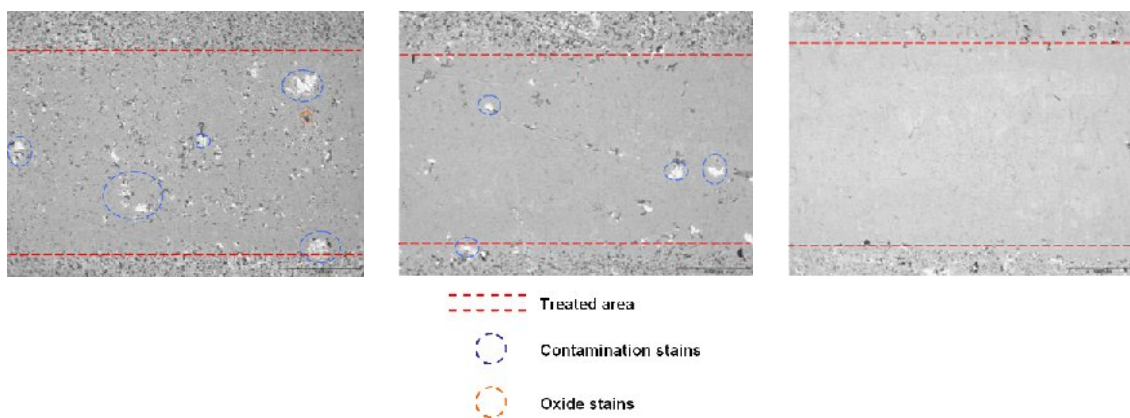


Fig. 2: SEM pictures (After one application (Left), after 2 applications (Center) and after 10 applications (Right)).

As we can see above, with the energetic level applied, ten applications are required to achieve a complete decontamination of the surface.

Results Obtained in Active Conditions

As mentioned at the beginning of this paragraph, two series of tests have been done on contaminated material.

For this first series, small pieces of a liquid waste tank have been tested. Those samples were covered with a thick layer of metallic oxides. The initial radiological conditions onto the surface were not completely homogenous (from 10 up to 1 000 counts per second).

Tests have been done in our TRIADE facility (near Tricastin NPP in the south of France) in automatic way using a robotic arm. The goal of these first tests series was essentially to demonstrate the efficiency of the LASER technology in presence of radioactivity.

To evaluate the efficiency, the LASER has been applied several times on the surface with different energy densities. After that, we calculated the percentage of decontamination and also decontamination factor. Some of the best results are listed in the table 1.

Table 1: Decontamination Results Obtain With LASER Process.

Sample #	# applications	Initial contamination (counts.s ⁻¹)	Final contamination (counts.s ⁻¹)	% of decontamination	DF
1	2	950	2	~ 100	> 450
2	4	1000	3	~ 100	> 300
3	4	960	3	~ 100	> 200
4	6	1000	3	~ 100	> 300
5	6	900	2	~ 100	450

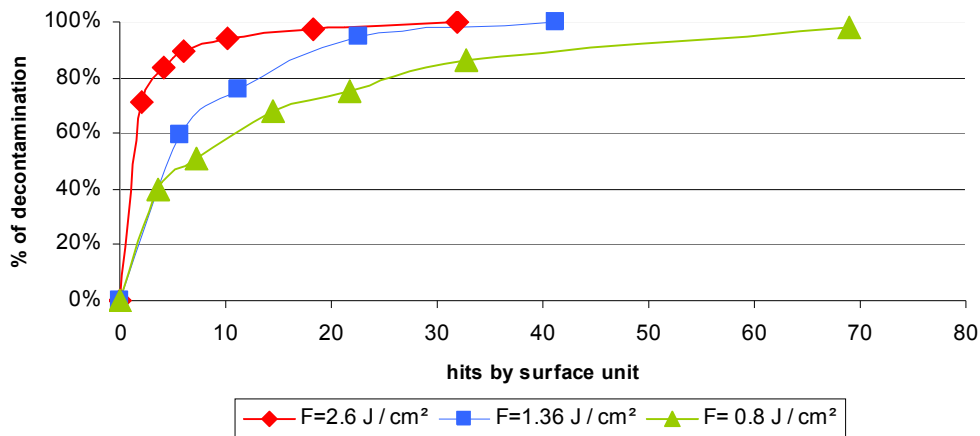
Figure 3 shows the results obtained on one sample.



Fig. 3: Sample before ablation (Left), Sample after partial ablation (Center), Sample after complete ablation on one part (Right).

The demonstration of such results is that we do not need to remove all the oxide layer in order to achieve decontamination. Indeed on the central figure, 100% of decontamination has been obtained even if a thin oxide layer remains. However, if for some reasons we need to remove the entirety of the oxide layer, we can do it as it is shown on the figure 3 (right).

The graphic 2 gives some information related to the percentage of decontamination in function of the hits number applied onto the surface.



Graphic 2: Influence of the energy density on the efficiency of the ablation process.

What can be learnt from this graphic is that, the higher the energy density, the faster the complete decontamination will be. Indeed for a density of 2.6 J / cm² we will need roughly 30 impacts versus more than 70 for a density of 0.8 J / cm².

For this second series of tests, metal pieces from La Hague have been tested. Those samples have spent a long time in a pool and were covered with a thin layer of grease.

The initial contamination was 400 counts per second in α and 7 000 in $\beta\gamma$.

Tests have been manually done in La Hague facility (AD1BDH decontamination workshop). The results confirmed those previously obtained at TRIADE. After applying the LASER during 5 minutes, the final radiological conditions have been measured at 4 counts per second in α contamination (DF = 100) and 140 in $\beta\gamma$ (DF = 50).

CONCLUSION

LASER technology appears to be an interesting one for the future of the D&D applications. As shown in this paper, the results in terms of efficiency are really promising and in many cases, higher than those obtained with conventional techniques. One of the most important advantages is that all those results have been obtained with no generation of secondary wastes such as abrasives, chemicals, or disks... Moreover, as mentioned in introduction, the LASER ablation process can be defined as a "dry" process. This technology does not produce any liquid waste (as it can be the case with chemical process or HP water process...).

Finally, the addition of a vacuum system allows to trap the contamination onto filters and thus avoiding any dissemination in the room where the process takes place.

The next step is going to be a commercial use in 2012 in one of the La Hague buildings.

REFERENCES

1. R. Bardtenschlager *et al.*, **Decommissioning of light water reactor nuclear power plants**, *Nucl. Eng. Des.*, 45, 1, **1978**.
2. C. V. Koch, R. Gruner, **Decontamination during decommissioning**, *Kerntechnik.*, 56, 372, **1991**.
3. H. Wille, H. Berthodolt, **System decontamination with CORD and decontamination for unrestricted release**, *Nucl. Power Performances and Safety.*, Vol 5, *Nuclear Fuel Cycle*, IAEA, CN-48/158, **1998**.
4. I. H. Plonski, F. Berevoianu and M. Toader, **Removal of contaminated oxide layer on carbon steel in hydrochloric acid disodium citrate solutions**, *J. Radioanal. Nucl. Chem.*, 185, 2, **1994**.
5. Z. Homonnay, A. Vértés, E. Kuzmann, K. Varga, P. Baradlai, G. Hirschberg, J. Schunk and P. Tilky, **Effects of AP-CITROX decontamination procedure on the surface oxide layer composition of stainless steel originating from the primary circuit of a VVER-Type nuclear reactor**, *J. Radioanal. Nucl. Chem.*, 246, 1, **2000**.

6. H. Wille, H. Berthodolt, F. Roumiguière, **Chemical decontamination with CORD-UV process: Principle and field experience**, *Proceedings of 4th Regional meeting nuclear energy in central Europe*, **1997**.
7. D. Noël *and al*, **Procédé de dissolution d'oxydes déposés sur un substrat métallique**, *patent FR 2 699 936 – A1*, **1992**.
8. N. Luis, K-MI Donald.,**Foam and gel methods for the decontamination of metallic surfaces**, *patent US 7,166,758 B2*, **2007**.
9. S. Bargues, F. Favier, J. L. Pascal, J. P. Lecourt. F. Damerval, **Organomineral decontamination and use thereof for surface decontamination**, *patent US 6,203,624 B1*, **2001**.
10. S. Faure, B. Fournel and P. Fuentes.,**Composition, mousse et procédé de decontamination de surfaces**, *patent FR 2841802*, **2002**.
- 11 K. Naessens, S. Boons, A. Van Hove, T. Coosemans, S. Verstuyft, H. Ottevaere, L. Vanwassenhove, P. Van Daele and R. Baets, **Excimer laser ablated U-groove alignment structure for optical fibre arrays**, *Proceedings Symposium IEEE/LEOS Benelux Chapter.*, Belgium, **1999**.

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