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Decontamination of metallic surfaces by nanosecond laser ablation

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► **To cite this version:**

L. Carvalho, W. Pacquentin, M. Tabarant, M. Dal, H. Maskrot, et al.. Decontamination of metallic surfaces by nanosecond laser ablation. International Conference On Laser Ablation 2017, Sep 2017, Marseille, France. International Conference On Laser Ablation 2017, 2017. hal-02417751

HAL Id: hal-02417751

<https://hal.science/hal-02417751>

Submitted on 18 Dec 2019

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Context & Aim of the study

The need of cleaning of equipment employed in the nuclear industry is growing and will be a major issue in the coming decades. To deal with such situation, the **decontamination of metallic equipment by laser ablation** is an advantageous technique that can provide the reduction of nuclear waste while minimizing the worker exposure. The efficiency of this process has already been proved but the previous studies showed the presence of a **residual contamination**. The goal of this study is therefore to determine the **limiting factors** of the technique by the comprehension of the physical phenomena involved.

- Study of the factors responsible for residual contamination
- Comprehension of ablation mechanisms induced by nanosecond regime using modelization
- Optimization of laser decontamination

Experimental protocol

Material

Stainless steel AISI 304L

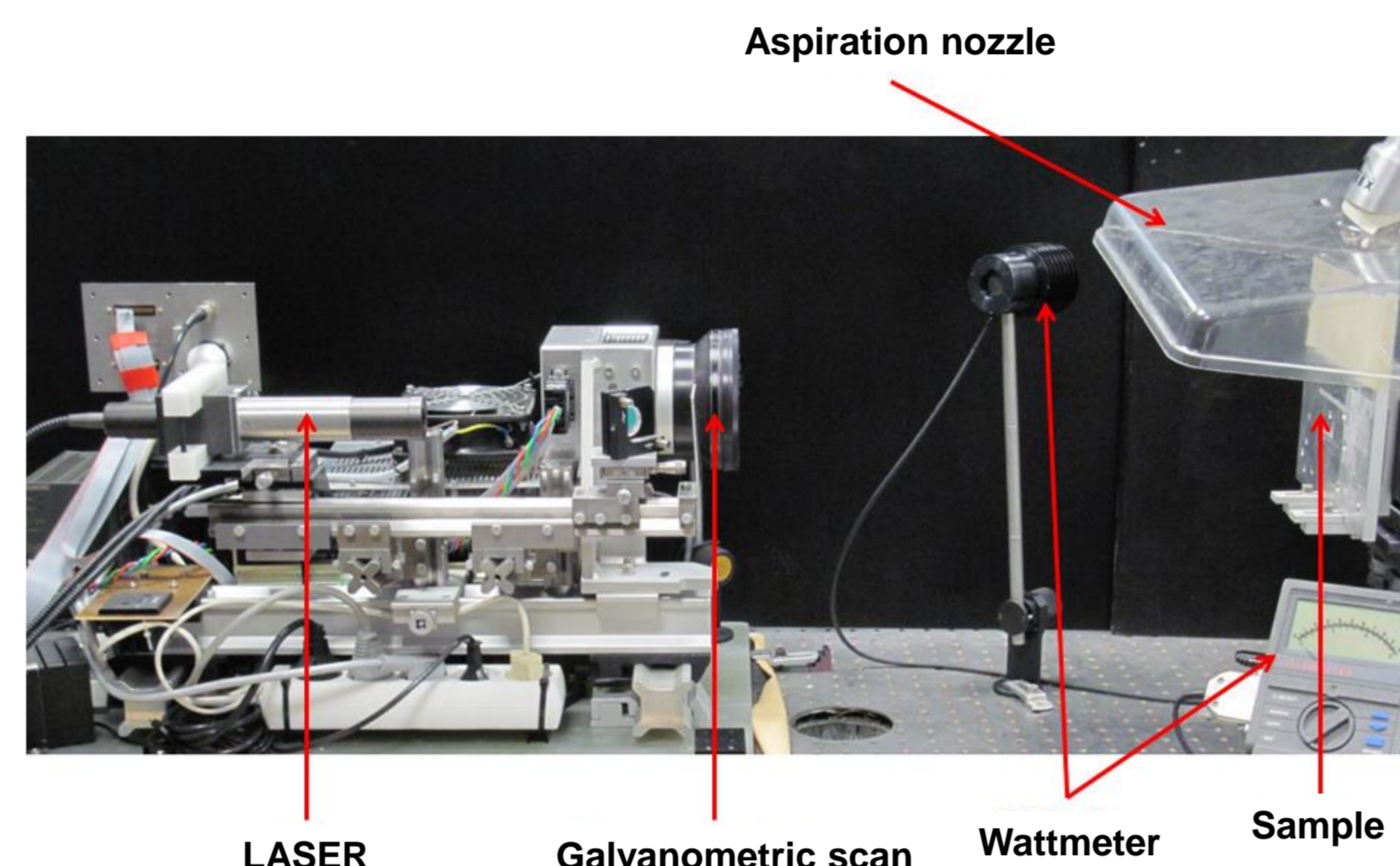
% weight	Fe	Cr	Ni	Mn	Si	Co	Mo	C
	Bal.	18.57	9.23	1.44	0.36	0.22	0.22	0.02

Sample preparation with tracing element incorporation

- Contamination by nebulization of $\text{Eu}(\text{NO}_3)_3$
- Fixation of Eu by laser melting
- Oxidation by thermal treatment in furnace

- Layer composition: $\text{Fe}_2\text{O}_3 + \text{Fe}_3\text{O}_4/\text{FeCr}_2\text{O}_4$
- Contamination in Eu in the whole volume
- Oxide layer thickness : from 100 nm to 2 μm

LASER set up



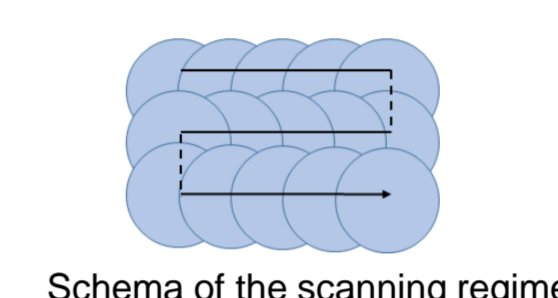
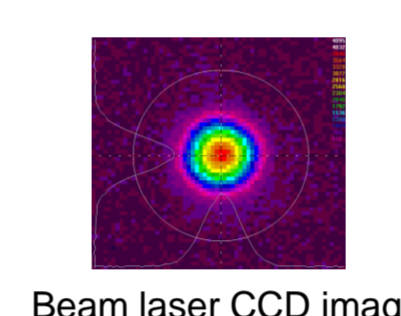
Yb fiber laser

IPG YLP-1/120/20
Wavelength: **1064 nm**
Gaussian beam ($M^2 = 2.1$)
Beam radius ($1/e^2$): 62 μm
Pulse duration: **110 ns**
Repetition rate: **20 kHz**
Mean power: **20 W**

- Scanning of the surface by consecutive laser pulses
- Collection of ablated matter
- Fast kinetics

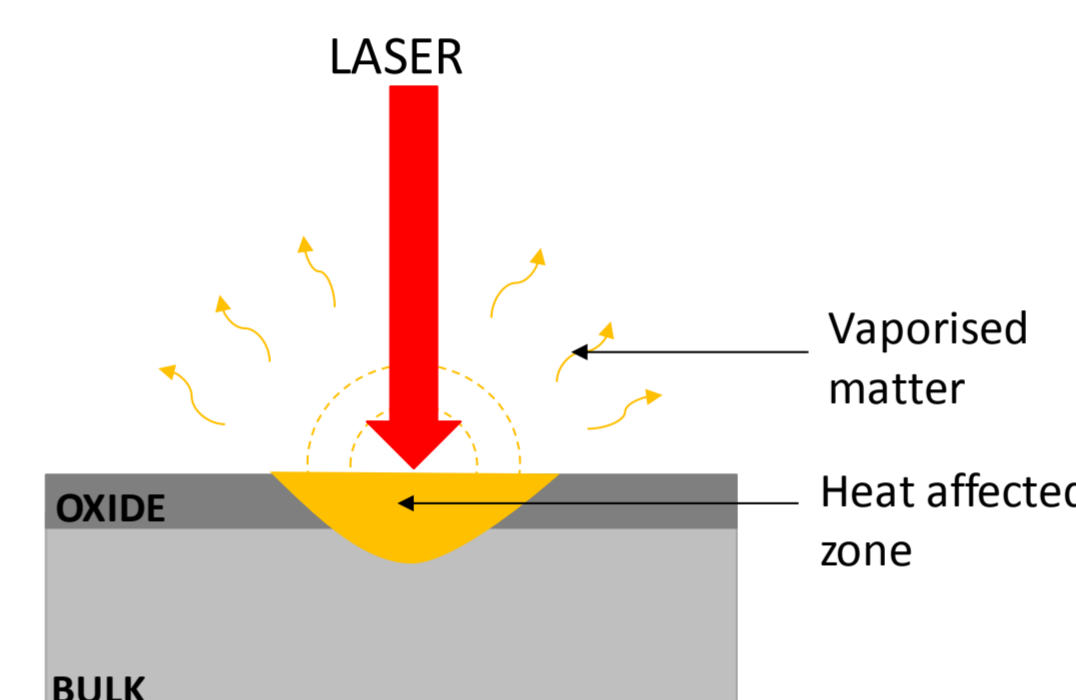
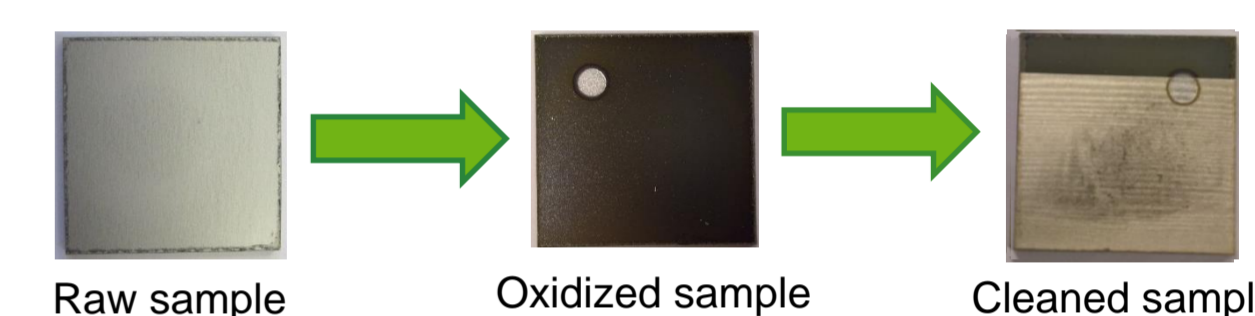
Experimental strategy

- Characterization of the ablated matter and the surface after cleaning
- Modelling of a laser interaction with metal surface



- Optimization of laser cleaning parameters

Parameter	Min	Max
Laser pulse fluence	5 J/cm ²	12 J/cm ²
Laser spot overlapping	60 %	90 %
Number of scan	1	10



Cleaning of oxidized metallic surface

Characterisation of the ablated matter

Collection of the aerosol on 0.45 μm nanopore filters

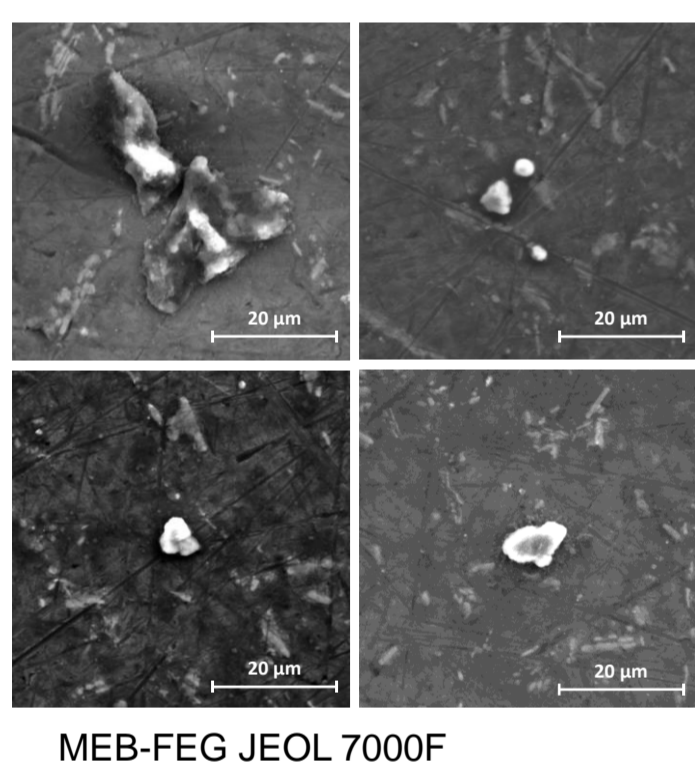
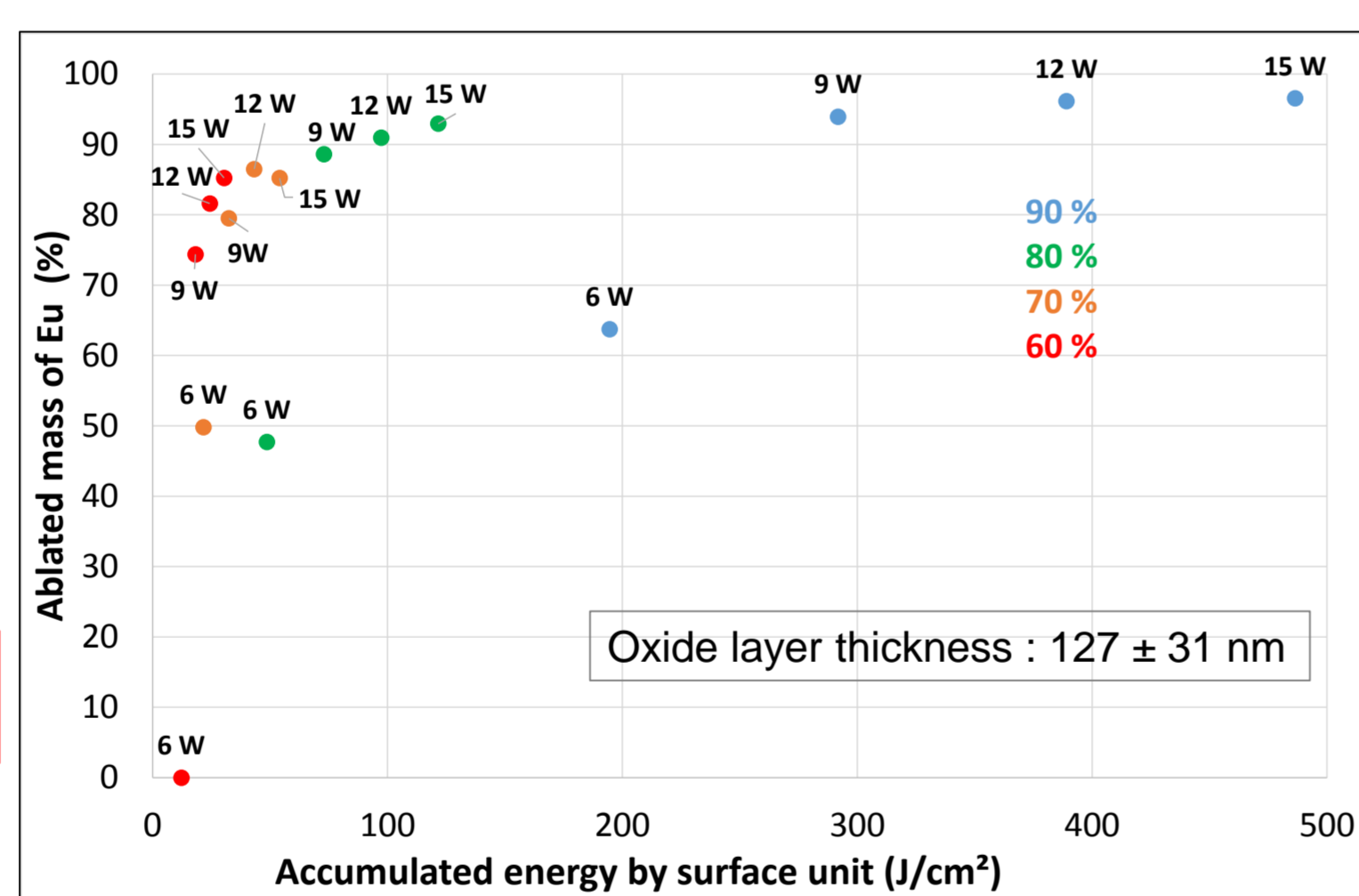


Image by Scanning Electron Microscopy (x1800) of collected aerosol

Observation of collected matter :

- Fragments of oxide (10 - 20 μm)
- Aerosols (1 - 5 μm)

→ Identification of ablation mechanisms



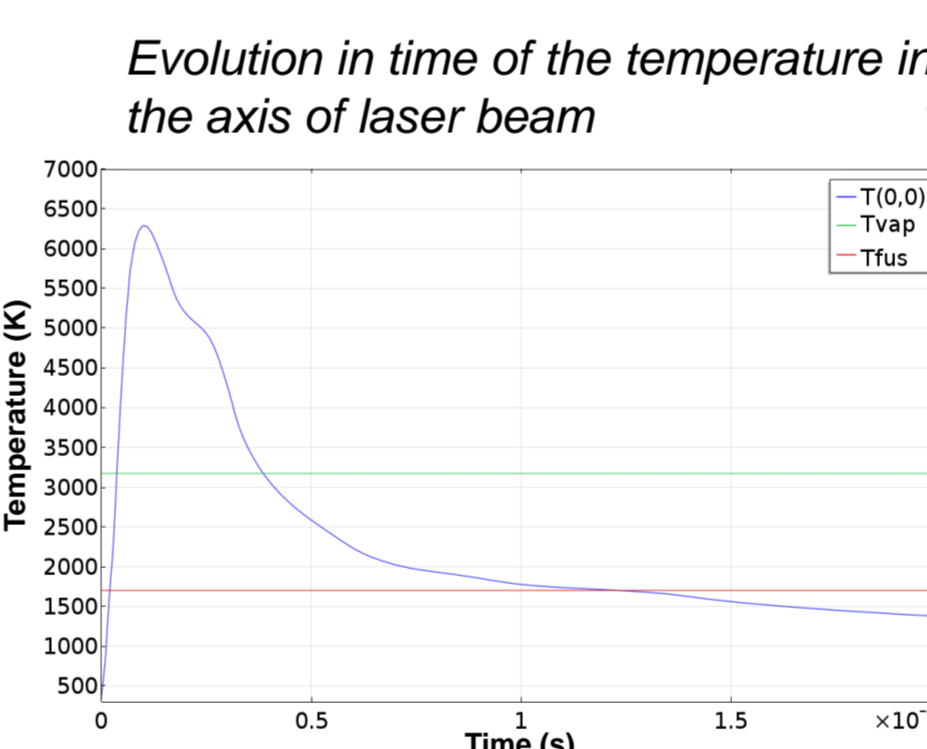
- ✓ Determination of a cleaning power threshold (**P = 6W**)
- ✓ Decontamination lower than 100 % after 1 scan → Residual contamination to be treated

Modelling of laser ablation

❖ Modelling of a single laser impact with COMSOL Multiphysics® :

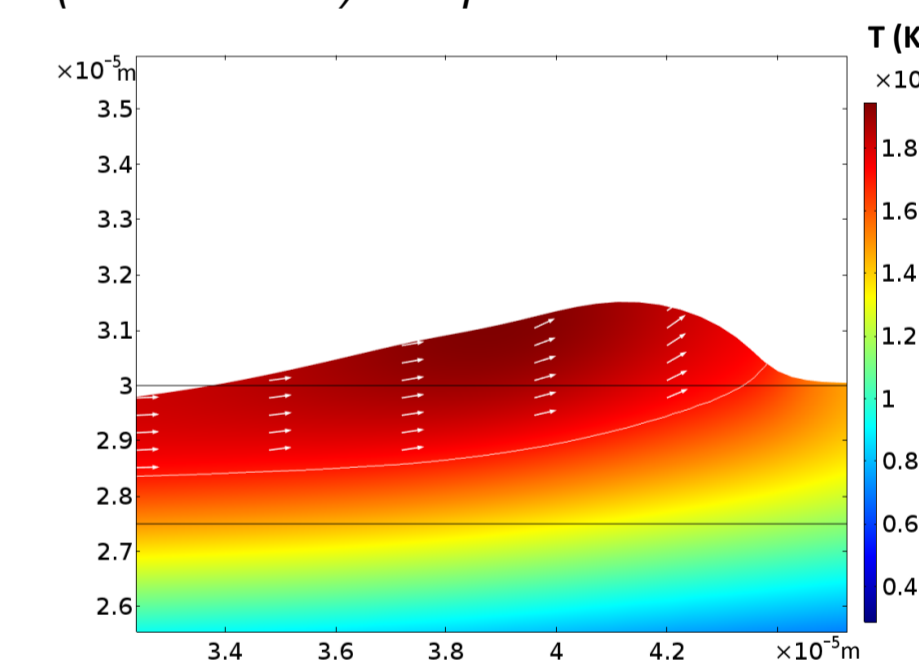
- Heat transfer and fluid flow
- 3D+t model
- Stainless steel AISI 304L

❖ Mechanisms of matter ejection :



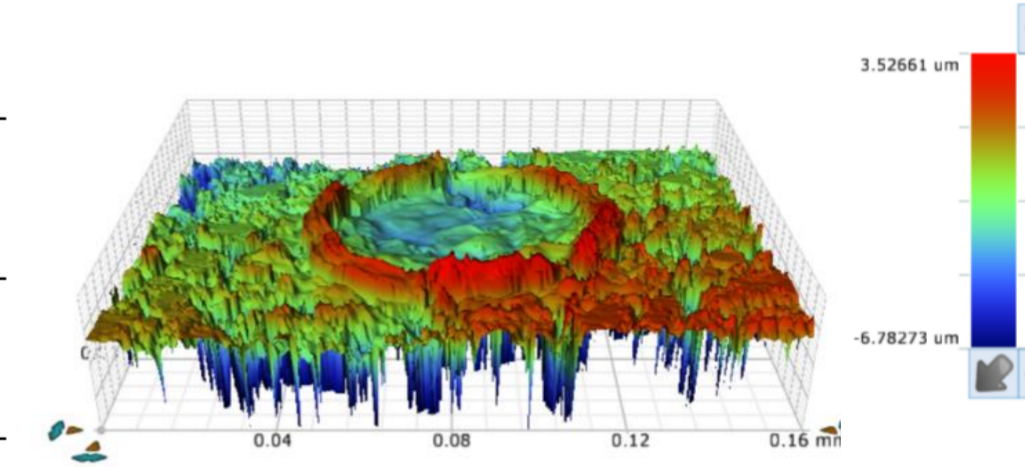
- Temperature arise and phase transition during the laser pulse (110 ns)
- Ejection of a melt pool (laminar flow)
- Formation of a ring at the solidification front

Temperature distribution (K) & velocity (white arrows) at 1 μs



❖ Comparison with experimental results

	Experimental	Simulation
Crater diameter (μm)	67.8 ± 1.1	72
Crater depth (μm)	0.93 ± 0.11	0.8
Ring height (μm)	1.14 ± 0.48	1.9



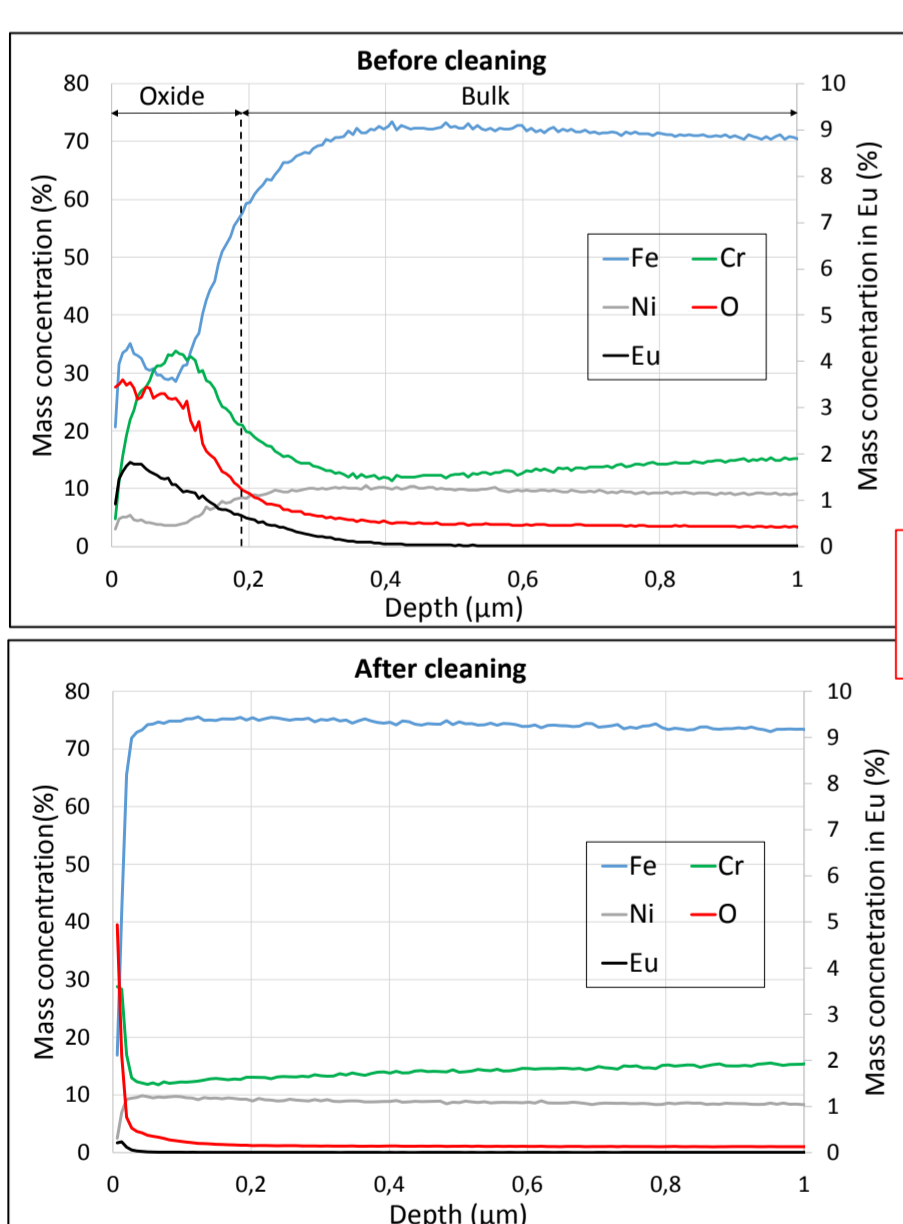
Results at laser fluence $F = 12 \text{ J/cm}^2$

→ Validation of the model for a single impact

Perspectives of modelization

- Modelization for a **submicrometric oxide layer** on a substrate to be closer to the studied oxide layer with a tracing Eu on stainless steel AISI 304L
- Modelization of **multi pulse interaction** to evaluate the effect of **heat accumulation** and of **liquid matter convection**
- Calculation of the **spatial distribution** of Eu to trace its **diffusion** in the bulk

Decontamination efficiency



Cleaning efficiency of **91 %** after 1 scan

Glow Discharge Mass Spectrometry profiles before and after decontamination (1 scan, 97.3 J/cm² accumulated fluence)

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Hypothesis of limiting factors

Penetration of Eu in the bulk during laser treatment

Entrapment of Eu in surface defects

Understanding of the influence of the accumulated energy during treatment and of the different thermal cycles

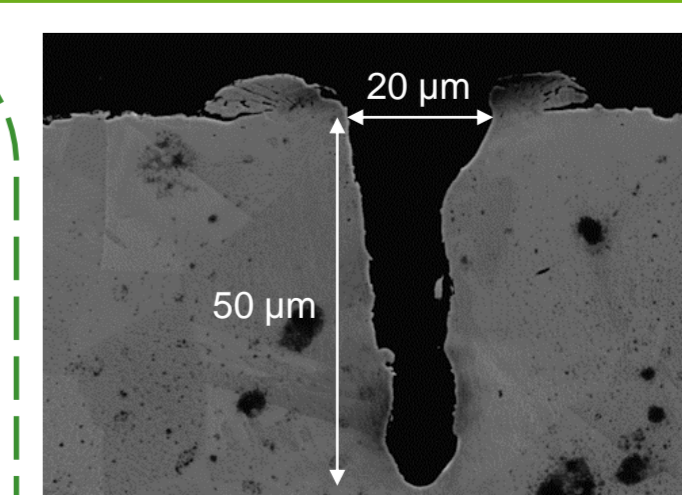
Decontamination of surface defects (cracks, etching)

Conclusions

- ✓ Development of an oxidation process with incorporation of Eu as a radionuclide simulant
- ✓ Decontamination of oxidized samples at a cleaning efficiency up to 90 %
- ✓ Experimental validation of the modelization of laser impact on a metallic surface

Perspectives

- Optimization of decontamination parameters
- Modelization of laser ablation by multi impact of metallic surface with an submicrometric oxide layer
- Cleaning test of surface with defects



Surface defects with controlled geometry

- Surface microetching by laser → Analysis at the bottom of the cracks
- Juxtaposition of removable polished plates → Analysis of matter and microstructure

