I.FAST

Innovation Fostering in Accelerator Science and Technology

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MILESTONE REPORT

Construction and operation of a High Temperature ALD system dedicated to SRF Cavities

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ABSTRACT

Particle accelerator construction and operation cost reduction is a crucial topic for future accelerators. This task aims at studying and developing new superconductors and structures to increase the performances of superconducting radio frequency cavities. To that end, we have built a custom deposition system that can handle various cavity sizes and shapes.

For more information on I.FAST, its partners and contributors please see https://ifast-project.eu/

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Delivery Slip

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Executive summary

Superconducting radio frequency (SRF) cavities are a cornerstone infrastructure of particle accelerators. Current bulk niobium technology is reaching its material intrinsic limits and new materials and structures are needed to both decrease the operation and construction cost while preserving or even improving their performances.

The approach underlying this task is to deposit superconductors with better properties than Nb as well as new superconducting multilayers that aim at increasing the accelerating gradient and the quality factor. We have selected atomic layer deposition (ALD) as a deposition technique of choice. ALD is known for its astonishing conformality on complex shapes objects with atomic level control of the thin film thickness and composition. ALD is therefore a bridge from coupons scale process optimisation to real scale particle accelerator objects, such as SRF cavities.

In this milestone report, we describe the features of a custom-built ALD system at CEA that can handle cavities of different sizes in a control atmosphere environment compatible with cavity materials and growth process parameters. The chemical delivery system and the vacuum oven deposition chamber are presented along with the methodology we developed to handle cavities in an accelerator-compatible environment (cleanliness, particle contaminations...).

The homemade ALD system have successfully passed the quality certification tests and three successful deposition runs on three niobium cavities have been achieved to date. Not only the CEA team were the first in Europe to deposit uniform thin films on SRF cavities by ALD but, for the first time worldwide, we have successfully deposited a superconducting multilayer on a niobium cavity.

These achievements pave the way to testing new thin film-based approaches for next generation of SRF cavities and particle accelerators.

1. Introduction

The development of alternative materials and structures for superconducting Radio Frequency cavities is a crucial aspect for future particle accelerators. In particular, construction and operation cost reduction is at the heart of contemporary energy and material restrictions preoccupations. This task fits into this broader goal and Work Package 9 task that aims at Cavity and accelerator cost reduction through Higher Gradient operation and new superconducting materials deposition with better superconducting properties than niobium. The selected deposition method: ALD is well known for its unequalled conformality on arbitrary complex shape objects that has been used in various fields at industrial scales (microelectronics, energy production and energy storage). Hence, this technics is ideal to bridge the gap between coupons scale research to real accelerator objects such as SRF cavities.

The construction of this homemade ALD deposition system will enable studying various approaches to reach this goal:

- Deposition of multilayers AlN/NbTiN on bulk Nb cavities to increase the maximum accelerating gradient.
- Deposition of Nano layers as dopant sources on bulk Nb cavities to increase the Quality factor.
- Study the deposition of ALD thin films directly on copper cavities prior to thin films depositions in order to : 1/ passivate the copper surface 2/ study its influence on Nb film growth quality and 3/ block thermal currents.

This homemade deposition system is design to handle various cavity sizes: from 6 GHz down to 0.65 GHz in niobium or copper as well as Quadrupole Resonators samples. The vacuum Oven enable heating up the cavity in a controlled atmosphere environment to prevent heavy oxidation. We designed various adaptors to easily handle different cavity flanges. The connections, deposition processes can be made by one trained person over a period of a week (typically 3 days) prior to shipping it back for subsequent treatments or deposition and testing at another laboratory.

We describe hereafter the various technical aspects of the custom-built deposition system: the chemical delivery system, the vacuum oven and the connections to the cavities as well as a typical process to avoid particulate contamination. Finally, the certification tests of the system are presented.

2. ALD SYSTEM

2.1 Chemical delivery system

The chemical delivery system (figure 1) include 9 precursors delivery lines:

- Four solid precursor delivery lines for which the chemicals are in powder form and held in stainless steel recipient called bubbler that can be heated up to 250°C to sublime partially the powder and deliver the precursor in vapor form. The temperature set point and uniformity are crucial parameters.
- One high temperature line that can be heated up to 450°C to deliver solid precursor with a high melting point.
- Two gas precursor lines.
- Two liquid precursor lines.

The delivery system is composed of 12 independently heated zones: 7 precursors zones, 2 tubes zones that brings the gases up to the reaction chamber located inside the oven, and 3 "exhaust" zones for the residual gas analyser (RGA), the connection cube zone and the below connected to the filter and the pump. The RGA is here to monitor in-situ the gas composition and the ALD processes.

It is important to maintain a temperature gradient from the precursor source to the reaction chamber in order to prevent powder condensation on the way that would contribute to an uncontrolled CVD regime.

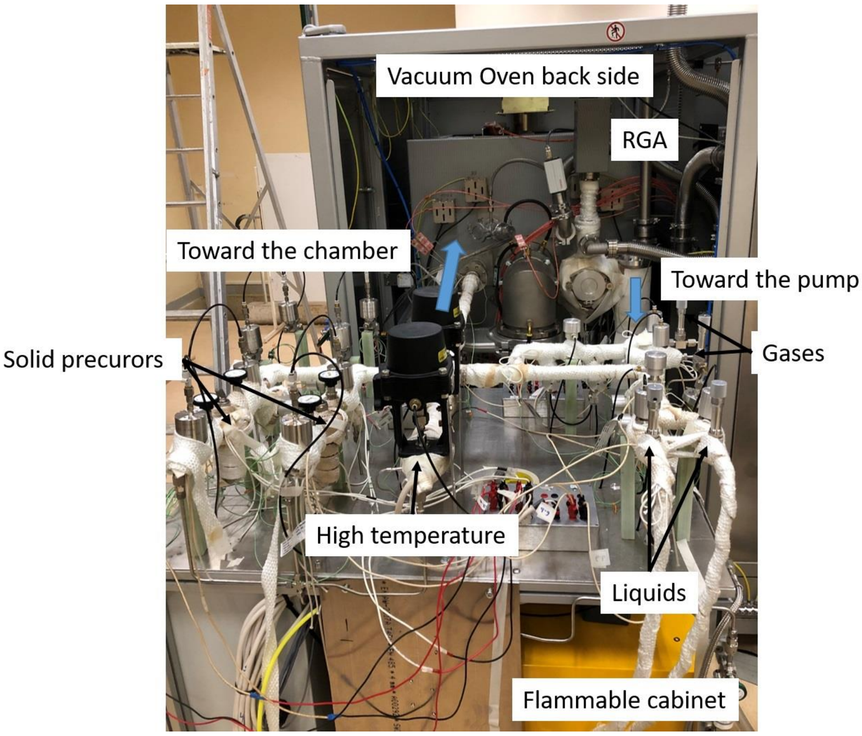

Figure 1: Chemical delivery system

The ALD system is typically at a pressure of 1 mbar under growth conditions and 350 sccm of inert gases flow. The inert gas can be switched between argon and nitrogen. Two mass flow controllers control their flow. The transport gases are five nines (99.999%) ultra-high purity gases further purified down to ppb level of H_2O, O_2, CO, CO_2 owing to gas purifiers located at the system entry line.

The RGA chamber is separated from the ALD lines by a pinhole and independently pumped by a turbo pump, reaching a vacuum level of 2.10^{-7}mbar during growth.

The ALD system can be vented and pumped back to growth or standby conditions via a large pneumatic valve located between the pump and the ALD gas lines.

The interface electronic is composed of a compactRIO from National Instrument to which is connected every thermocouples (40), heating output to the 12 heating zones, two mass flow controllers, two pressure sensors, and 20 pneumatic valves actuators. The RGA is directly connected to the computer via RS232 protocol.

2.2 The vacuum Oven and adaptors

The Vacuum Oven from the company Nabertherm (figure 2) can go up to 650°C under vacuum and 980°C under inert gaz. The vacuum level at 450°C is typically $1.0\ 10^{-6}$ mbar and pumped by a CF200 flange turbo. The retort volume hosting the reaction chamber (cavities, large tubes…) is 50 cm in diameter and 100 cm long. The custom part of this oven is located on the backside of the oven, close to the chemical delivery system, and consist in two feedthroughs through which the gases are injected to the chamber and exhausted to the pump.

Figure 2: front views of the vacuum Oven with retort door closed or open.

Inside the retort, adaptors connect the cavity or large chambers to the ¾" VCR ALD delivery lines (figure 3). Two types of adaptors are currently at disposition: CF100 flanges for large tube like chamber and 1.3 GHz cooper cavities and DESY-tesla type flanges for Nb 1.3 GHz flanges. We are in the process of making larger flanges for 0.7 GHz cavities as well.

The adaptors are conic in order to prevent abrupt diameter change along the gas path that could create local low gas speeds and hence long purges. Ten thermocouples are at disposition inside the retort to measure temperature uniformity along the chamber/cavity. At 450°C, the temperature uniformity is ± 1.5 °C. The chambers or cavities are maintained in position by titanium supports resting on a titanium plate.

The Vacuum Oven program enable multiple ramps, ramp rate and set points to build complex heating profiles. Several profile are saved for different growth conditions.

Various parts of the vacuum oven are water cooled down by a cold unit located in the basement under the laboratory.

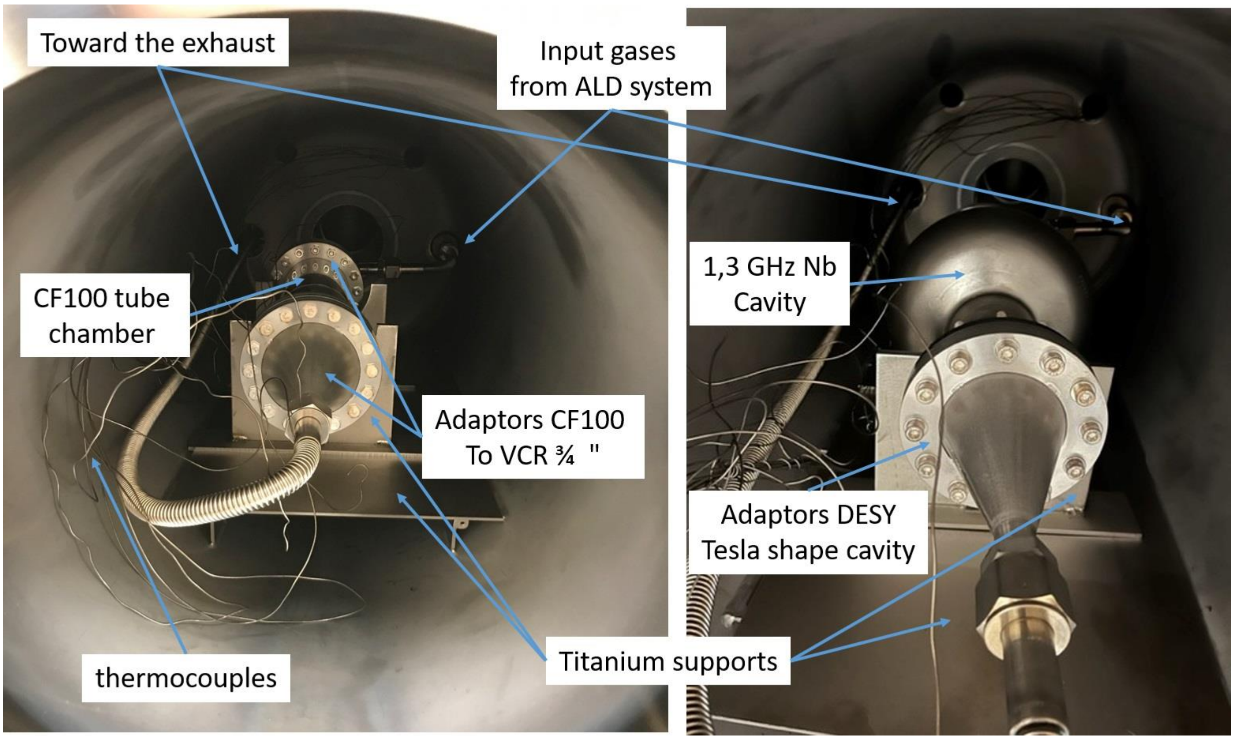

Figure 3: chamber (left) and cavity (right) installed inside the oven retort with adaptors.

The procedure to coat a cavity starts with the clean room assembly of the adaptors on the cavity, after the HPR. The cavity and adaptors assembly is closed in the clean room and then transported to the laboratory. This assembly is then connected inside the retort to the ALD system via the VCR fittings. A Helium leak check is done on the connection prior to closing the oven door. The pumping and heating to the set point takes about six hrs to reach good temperature profile uniformity along the cavity/ALD lines. After the deposition process, the cool down of the retort takes about another 6 hrs in accelerated mode owing to forced air flow outside the oven through the resistance space.

3. Control program

The LabVIEW control program was written by Yannick Le Noa from the Department of engineering systems (DIS) following the structures and instructions I wrote (figure 4). The program communicate with the compactRIO and controls the ALD chemical delivery system. It has been designed to be easy and intuitive to manipulate. It enables the redaction of recipes and save various process parameters during the deposition time.

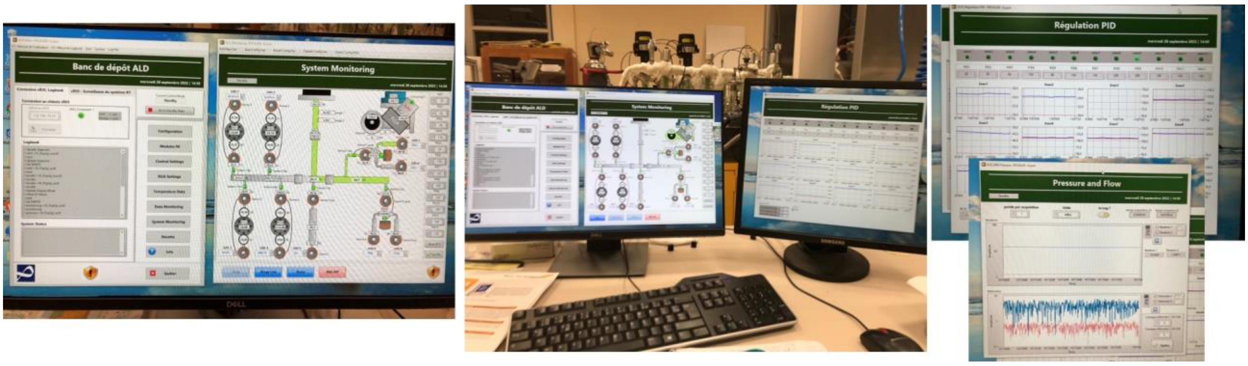

Figure 4: Labview control program. Left: schematic of the ALD precursor delivery system where is displayed the zones temperature and pneumatic valves status. Right: Pressure and flow control windows being monitored during an ALD process.

The program has different modes from experts to beginners that enable access to different level of parameter control. Repeated and overseen training by the Principal Investigator are required to access the expert level. The program has safe guard limits on all process parameters (flow, temperature, pressure) that trigger a stop of the growth process if over reached.

The major improvements compared to the small scale, previously built, ALD reactor are:

- Simplification of the inert gases delivery system that only require 2 mass flow controllers instead of one per chemical delivery lines (here 7), reducing significantly the price (1k€/mass flow)
- Increased speed of monitoring and recording processes.
- The recipe is deported to the compactRIO in order to continue the growth in case of computer shut down (system updates that can happen anytime and give you 4 hours to act prior to reboot) that can happen during long overnight or several days growth.

It took numerous back and forth (about 1 year) between program testing in real conditions and programming to converge to a stable state and clear out most bugs and errors. Some features can be improved still but the program is now in a stable working condition.

4. System certification

The tube like chamber 100 mm in diameter was used to test the ALD system. Samples are positioned inside and along the 50 cm tube. Simple Al2O3 recipes at 250°C were tested at first and the film thickness homogeneity were measured ex-situ by X-ray reflectivity (XRR). The results show a thickness homogeneity on the targeted 50 nm thick Al2O3 films of 1.5%. Using the same methodology, we subsequently tested TiN deposition and more complicated multilayer nitride recipes: AlN-NbTiN at 450°C in the tube like chamber. The film thickness uniformity ranged between 2 to 3% along the chamber on 50 to 100 nm thick films. The electronic properties (room temperature resistivity, superconducting transitions…), structures and chemical compositions were quite homogeneous as well and reproduced the values obtained on the other, research scale ALD system.

We replaced the tube like chamber by niobium cavities and did three kind of depositions:

1- One TiN 2 nm thick deposition on 10 nm thick Al2O3 at respectively 450°C and 250°C.
2- One multilayer: 7 nm thick AlN and 50 nm thick NbTiN at 450°C.
3- One NbN 5 nm thick deposition.

For depositions 1 and 3 we cannot expect colour changes inside of the cavity as the films are two thin. For deposition 2 however (figure 5), the changes inside of the cavity were clearly visible and the films looks quite homogeneous.

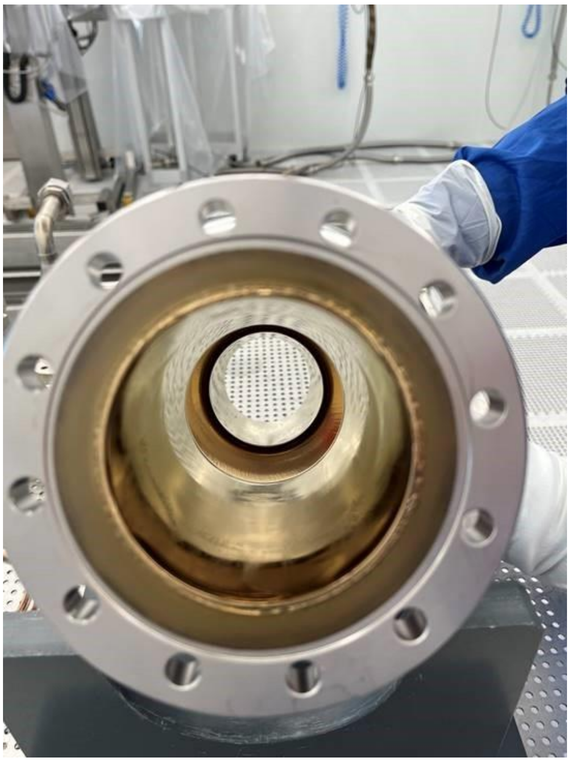

Figure 5: visual inspection of a 1.3 GHz Nb cavity after ALD synthesis of a multilayer: AlN/NbTiN.

5. FUTURE PLANS / CONCLUSION / RELATION TO OTHER ARIES WORK

Now can start the hard part of the work that involve tuning the films (thickness and compositions) and post treatments (temperature of post annealing) parameters on SRF cavities. Three main directions will be studied:

1- The multilayers of nitrides: AlN/NbTiN. For which the thickness of the NbTiN layer will be varied to optimize its screening efficiency. Various post annealing conditions under vacuum will be studied as well.
2- A new doping approach will be investigated that uses nitrides layers as a well-controlled source of dopants and surface layer engineering. Various post annealing conditions under vacuum will be studied. The goal is to avoid post electropolishing.
3- ALD deposition on Cooper cavities of insulating layers as seed layers and thermo-current barriers for Nb films deposition (collaboration with CERN).

In the task 9.4 work description, cavities will be supplied by INFN (6 GHz and eventually cooper 1.3 GHz in task 9.2) and tested at INFN (6 GHz) and CEA (1.3 GHz). We have also a QPR from task 9.6 ready to be coated at CEA and to be tested at HZB.

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