FURNACE N_2 DOPING TREATMENTS AT FERMILAB *

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Abstract

The Fermilab SRF group regularly performs Nitrogen (N_2) doping heat treatments on superconducting cavities in order to improve their Radio Frequency (RF) performances. This paper describes the set up and operations of the Fermilab vacuum furnaces, with a major focus on the implementation and execution of the N_2 doping recipe. The cavity preparation will be presented, N_2 doping recipes will be analyzed and heat treatment data will be reported in the form of plot showing temperature, total pressure and partial pressures over time. Finally possible upgrades and improvements of the furnace and the N_2 doping process are discussed.

INTRODUCTION

High temperature Nitrogen (N_2) doping baking activities for Superconducting Radio Frequency (SRF) cavities are performed on a regular basis in the SRF Department at Fermilab as a mean to improve Q_0 at medium gradients. Q_0 values increase as much as a factor of three at 18 MV/m accelerating gradient [1]. The SRF group owns two vacuum furnaces similar in design but of different size in order to accommodate various types of resonators. This paper will describe in detail how the N_2 doping process is executed, with a particular focus on the furnace operation and how a low pressure atmosphere of N_2 can be maintained in the furnace; finally bake data will be discussed.

FERMILAB VACUUM FURNACES

Two vacuum furnaces manufactured by TM Vacuum [2] are used for baking SRF cavities: the smaller one is represented in Figure 1. They are known as *Small TM* and *Big TM* and are comparable in their characteristics and design, but have a different working volume. Both have a base vacuum of 1×10^{-8} Torr and their maximum operating temperature is 1000°C (under vacuum or backfilled to 1 Torr). Heating is achieved by means of low voltage 2 inches molybdenum (Mo) strips and the hot zone is thermally insulated by six layers of Mo reflective shields, backed by a stainless steel containment. The vacuum chamber and door are double walled for the circulation of cooling water. The chamber is evacuated by a dry roughing pump and high vacuum is achieved by 2 cryopumps. A SRS RGA100 [3] Residual Gas Analyzer (RGA) is mounted on each system and is used to qualitatively follow the evolution the the vacuum atmosphere during a bake. If the pressure raises above 5.5×10^{-5} Torr during a bake cycle, the furnace goes into a *hold* mode: it maintains a constant temperature and lets the pressure drop down below 5.5×10^{-6} Torr; after that, the bake cycle is continued. The furnace also allows for the introduction of a process gas with the flow manually regulated by a metering valve. Once the bake is completed, the chamber is backfilled to atmospheric pressure with boil-off Argon or Nitrogen.

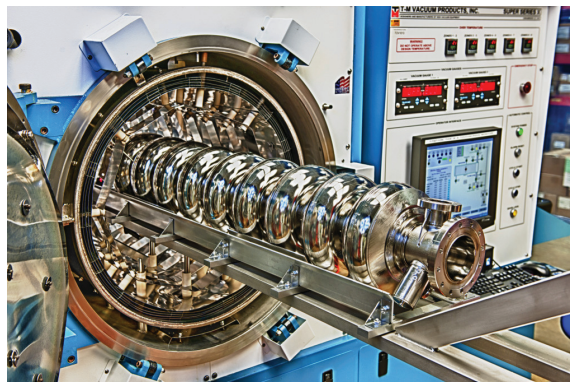

Figure 1: Super Series Vacuum Furnace SS 12/72-10 MCX from TM Vacuum, *Small TM*. Hot zone 12 x 12 x 72 inches. Mainly used for 1.3 GHz single and 9-cell cavities.

Gas Bleed Setup

Fermilab furnaces allow for the introduction of a process gas into the chamber at any temperature.

Figure 2 shows a schematic of the gas bleed setup. The metering valve (Swagelock SS-4MG [4]) is manually adjusted, while the isolation control valve is operated by the furnace PLC: its status (open or closed) is reported on the computer user interface in Figure 3 and it is identified by the name *bleed valve*. The metering valve is not leak tight which means that when the control valve is closed there will always be a trapped volume full of the process gas (N_2 for SRF cavities) between the two valves as shown in Figure 2. This volume of gas will be injected in the furnace as soon as the isolation control valve opens. After that, while the control valve stays open, the gas continues to flow in the chamber at the rate that is set by the metering valve. The optimal position of the metering valve to ensure the desired average chamber pressure of 25±2 mTorr was determined by trial and error during the initial development of the doping recipe.

A more precise setup can be obtained substituting the manual metering valve with a Mass Flow Controller (MFC) which allows a more accurate control of the flow. The size of the MFC orifice should be designed to rapidly bring the furnace from Ultra High Vacuum (UHV) to 25 mTorr and to maintain this average pressure compensating for the N_2 absorbed by the cavity. A MFC could also allow for detailed studies on the N_2 absorption of resonators and it would be

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possible to create accurate models for the process and make predictions on the flow required when baking more cavities simultaneously.

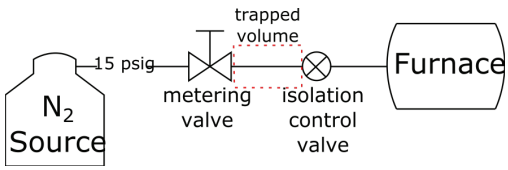

Figure 2: Gas bleed line sketch.

User Interface

The furnace is be operated by a computer and controlled by a PLC with the software interface is shown in Figure 3; from here the user can monitor the furnace status, actuate valves, turn on and off pumps and heating elements. One can also create a bake recipe and start a heating cycle. All the pneumatic valves are represented by a green circle: bright green indicates an open valve, (e.g. *high vacuum valves* are open in Figure 3), dark green is for a closed valve (e.g. *vent valve* is closed in Figure 3). Pressure sensors report the pressures measured in different parts of the furnace. The software is the same for both furnaces with the only difference that for the *Big TM* it is possible to keep the *rough valve* closed while the *bleed valve* is opened (i.e. during the N_2 doping). The *RGA isolation valve* is generally open and the RGA measures the vacuum quality; during the gas bleed, when the pressure raises above 10 mTorr, the *RGA isolation valve* closes automatically to protect the RGA. When automatic bake cycle is running, the *RGA isolation valve* is the only valve that can be operated manually, but only once the pressure drops below 10^{-6} Torr.

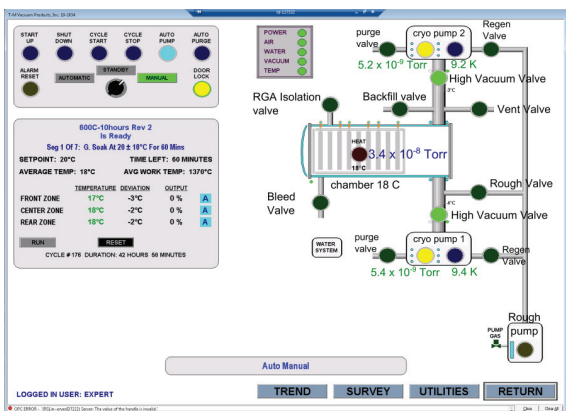

Figure 3: *RSView* software user interface for the Fermilab vacuum furnaces. Bright green indicates an open valve and dark green is for closed valves.

At the end of the bake cycle, when the furnace is at room temperature, all the valves will be closed except for the *vent valve* which allows boil-off N_2 inside the chamber and brings it to atmospheric pressure.

CAVITY PREPARATION

Before baking, all cavities receive ultrasonic degreasing with a 2 % Liquinox solution at 60°C, High Pressure Rinsing (HPR) and are dried in a cleanroom. Cavities are supported in the furnace chamber through the Mo hearth shown in Figure 4. A Niobium liner is placed between the cavity and the Mo hearth so that the resonator surface is only in contact with Nb avoiding any potential for Mo inclusion in the superconducting Nb. Survey thermocouples are also connected to the two resonator beampipes to measure the work piece temperature. The beampipe and other ports are covered with Nb caps which were originally introduced to avoid the light electropolishing (EP) step after the bake. In fact, during a bake, the Titanium (Ti) contained in the NbTi flanges will outgas due to its high vapor pressure and will deposit on the colder surfaces (e.g. heaters when cooling down). During the warmup in following bake cycles, the Ti previously deposited on the heaters will evaporate and migrate to colder surfaces (cavity). The caps prevent any line of sight between the inside of the resonator and the heaters avoiding any Ti deposition on the inner RF surface of the cavity and therefore ideally avoid the light EP step. However, light EP is still performed after a bake to remove the Nb nitrides that grow on the cavity surface after the N_2 doping. Figure 4 shows the typical setup of a cavity inside the furnace ready for a heat treatment. It is possible to identify the Mo hearth, the Nb liner and the beam pipe caps.

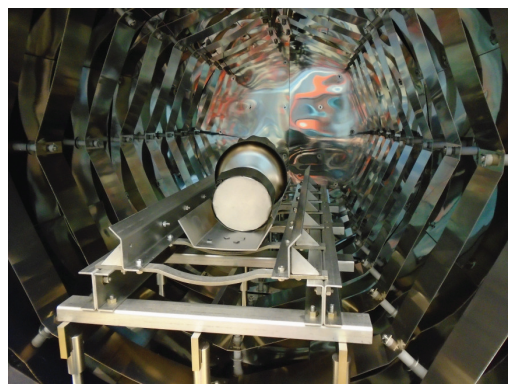

Figure 4: Typical cavity set up inside the vacuum furnace. This is a 1.3 GHz single cell cavity baked with 4 Nb samples *Big TM*.

NITROGEN DOPING RECIPE

The N_2 doping recipe is a slight modification of the standard H_2 degassing bake recipe implemented high performance RF cavities [1]. N_2 doping can improve Q_0 by a factor of two or greater at a gradient of 18 MV/m over the standard processing recipe developed for XFEL or ILC (Q_0 at 18 MV/m $\approx 3\times10^{10}$ for a N_2 doped cavity versus a Q_0 of $\approx 1.5\times10^{10}$ at the same accelerating gradient for a standard H_2 degassing recipe) [1], [5].

Different N_2 doping recipes have been developed during the initial R&D phase and generally a two step N_2 diffusion segment is added to the standard 800°C bake [5], [6]. The heat treatment sequence is the following:

1. pump-down to base vacuum; the bake is generally started when the pressure in the chamber is below 10^{-7} Torr.
2. leak rate test, to ensure that the vacuum furnace is properly sealed and there are no leaks that will damage the cavity and the furnace when at high temperature.
3. heating, ramp up to 800°C at a rate of 3°C/minute.
4. 800°C soak for 3 hours to remove hydrogen (H_2) accumulated during the bulk EP.
5. N_2 doping at 800°C.
6. 800°C annealing in Ultra High Vacuum (UHV).
7. Cooldown.

High values of Q_0 have also been obtained with doping recipes omitting the 6^{th} step [7]. Figure 5 is a plot of a typical doping bake highlighting all the segments described above.

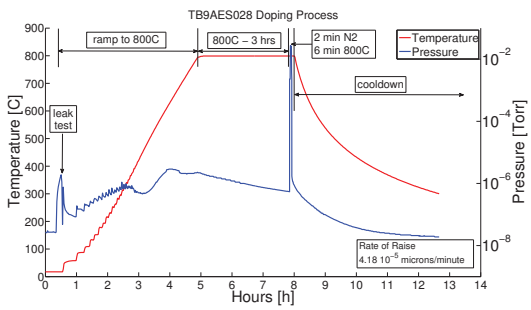

Figure 5: Process of a N_2 doping bake. The *High Q_0 recipe* is plotted here.

N_2 Doping Execution

When injecting gas at high temperature (e.g. N_2 for SRF cavities), the furnace PLC performs the following actions:

1. Close *high vacuum valves*, i.e. the cryo pumps are isolated from the vacuum chamber.
2. Maintain a temperature of 800°C (or other process temperature).
3. Close the *RGA isolation valve* to prevent any damage to the RGA when operating at high pressures; the RGA is manually shut down.
4. Open the *bleed valve* to introduce N_2 gas in the chamber.
5. Maintain the chamber at an average pressure of 25 mTorr for a specified amount of time.
6. Close the *bleed valve* and open the *high vacuum valves*.
7. Pump down to UHV.

While introducing N_2, the chamber has to be maintained at an average pressure of (25± 2 mTorr) as this value has been determined to be the optimal parameter. In both furnaces, the average chamber pressure is a result of the injection of N_2, N_2 absorption of the cavity and pumping action of the roughing pump; the furnace feedback control loop actuates the *bleed* and *rough valves* ensuring the required pressure range. *Big TM* can also be set up not to have active pumping during the N_2 bleed segment (i.e. the *rough valve* is maintained closed); in this case the average pressure is a combined result of the cavity absorption and the PLC control logic opening and closing the *bleed valve*.

High Q_0 Recipe

Among all the N_2 recipes developed at FNAL, one has proven to be the optimal in terms of Q_0 and quench performances and has also been chosen for all the LCLS-II cavities; this is known as the *High Q_0 recipe* and consists of 2 minutes N_2 doping at 800°C and 6 minutes annealing at 800°C in UHV [5], [7]. Bake plots of a *High Q_0 recipe* are reported in Figures 5 and 6 and is discussed in the following section.

BAKE RESULTS

This section reports bake results in the form of plots that will be thoroughly discussed and explained. Figures 5, 6 and 7 show typical plots of a *High Q_0 recipe* implemented a 9-cell 1.3 GHz cavity.

A doping heat treatment can be considered successful if H_2 is removed from the cavity and the cavity is doped with N_2; this can be assessed by looking at the mass spectrum measured by the RGA together with the evolution of pressure and temperature.

Tests at Fermilab show that no significant difference in cavity performance has been identified between cycles that had active pumping during the N_2 injection and those that did not [7].

N_2 doping may also be affected by the beam pipe caps design (sheet or foil) as their tightness may slightly change the N_2 pressure inside the cavity due to gas conductance. This effect has not been observed on 1.3 GHz cavities, but it is under investigation for 650 MHz resonators which were baked with different caps than those used for 1.3 GHz.

Pressure Plot

The chart in Figure 6 is the bake plot of the *High Q_0 recipe* and presents the evolution of temperature, total pressure and partial pressure of gas species of interest. In UHV, the total pressure in the chamber is measured by a cold cathode gage which is calibrated using N_2 and may not be able to accurately measure the H_2 contribution to the total pressure. This explains why H_2 partial pressure measured by the RGA in Figure 6 is higher than the total pressure in the chamber.

800C Mass Spectrum

The spectrum in Figure 7 shows the gas composition inside the vacuum chamber. It is possible to follow the evolution of the vacuum atmosphere throughout the bake. Here, one can easily identify the presence of hydrogen (H_2), water (H_2O), nitrogen (N_2), Oxygen (O_2) and carbon dioxide (CO_2), typical of a system that was previously opened to air and that contains a superconducting resonator. During the N_2 injection at a pressure of ≈ 25 mTorr, the RGA is

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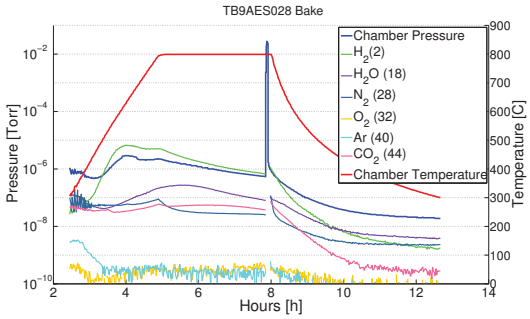

Figure 6: Bake plot of a N_2 doping bake (*High Q_0 recipe*), the evolution of the gas species of interest is monitored over time.

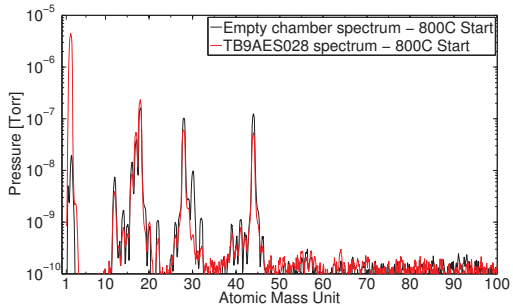

Figure 8: Comparison of the RGA spectrum at the beginning of the 800C 3 hours soak between an empty chamber and the bake of TB9AES028.

shut down to avoid damaging the filament. This implies that there is no measure of the gas components in the vacuum atmosphere. However, the cold cathode and thermocouple gauge are still measuring the total pressure in the chamber which can be assumed to be approximately equal to the partial pressure of the N_2 (the other components are order of magnitude lower so their contribution to the total pressure can be neglected).

Specifically, Figure 7 shows the spectrum at the beginning and end of the 800°C soak. The previous presence of N_2 (due to doping) can be detected by the higher N_2 at the end of the 800°C soak ($amu_{N_2} = 28$).

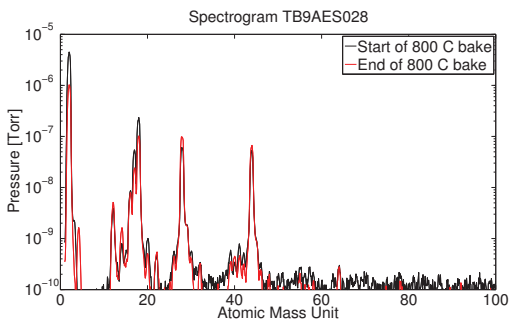

Figure 7: Spectrum at the beginning and end of the 800C segment. The end of the bake is at the end of the 6 minutes with N_2.

Empty Chamber vs Cavity

A comparison between the spectrum of an empty run and a cavity bake is sown in Figure 8 so it is possible to see what gases or contaminants are introduced in the chamber when baking a resonator. Both spectrum are very similar except for the higher partial pressure of H_2 that is measured from the RGA. There is no presence of gases with higher amu (typically hydrocarbons) which indicates that the cavity is clean and no contaminants are introduced in the furnace.

H_2 Outgassing

A large quantity of H_2 has been found in the Nb of SRF cavities that received tumbling before the heat treatment [8]. H_2 starts outgassing at ≈ 500°C and increases significantly the pressure in the furnace which enters the safe *hold* mode. Here, the temperature is maintained constant until the pressure decreases below 5.5×10^{-6} Torr, then the bake cycle is continued regularly and the N_2 doping is executed. Figure 9 shows the typical bake cycle of a tumbled cavity, with the classic plateau at ≈ 500°C. This behavior is not observed for cavities that did not receive tumbling.

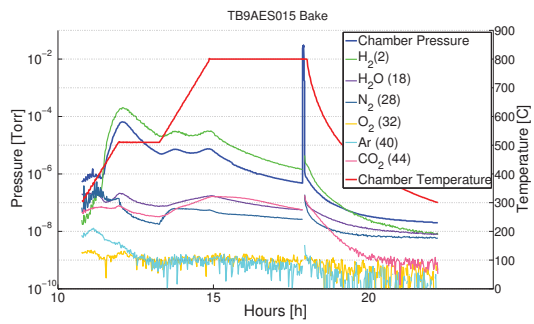

Figure 9: Example of the bake process with excessive H_2 outgassing due to tumbling.

Vacuum Atmosphere Evolution

Figures 10 and 11 show the evolution of the main gases in the furnace before and after the doping section of two cavities baked in the two different furnaces. The cavities received the same *High Q_0 recipe* and both had active pumping during the doping segment. TB9AES028 was baked in *Big TM* while TB9AES024 in *Small TM*. When the RGA is turned on again after the N_2 injection, the partial pressure of CO_2 for TB9AES028 is higher than that for TB9AES024. This difference between the two furnaces is always present, however tests performed on cavities have proven that the higher CO_2 partial pressure is not a determining factor for quench and high Q_0 performances.

SAMPLE ANALYSES

Small Nb witness samples have been baked together with cavities. The samples were placed on the Nb liner next to the cavity being baked, as shown in Figure 4. Scanning Electron

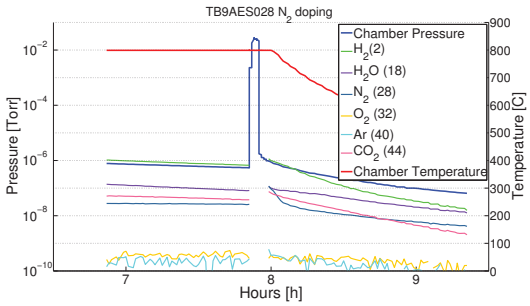

Figure 10: TB9AES028 N_2 doping section of the heat treatment. Cavity baked in *Big TM*.

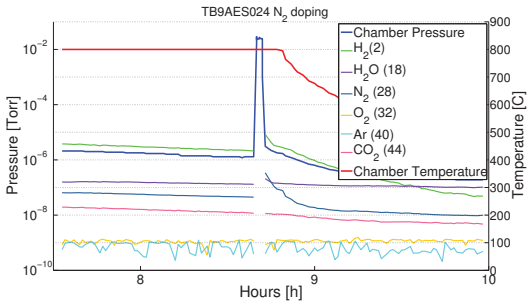

Figure 11: TB9AES024 N_2 doping section of the heat treatment. Cavity baked in *Small TM*.

Microscope (SEM) images have shown nitride inclusions on the Nb sample surface that increase their size with heavier doping [9].

SIMS Analyses

The concentration of C, N, H, O has been investigated using the Secondary Ion Mass Spectrometry (SIMS) technique. Figure 12 plots the N_2 concentration versus depth for four different samples. In the first μm poorly superconducting Nb nitrides are formed. Deeper in the bulk, superconducting Nb is found with interstitial N_2. Therefore, in order to obtain good performances after N_2 doping, few microns of material need to be removed from the surface. Figure 12 shows is a higher concentration of N_2 for samples more heavily doped, and this is in accordance with the expectations. Sample H from Figure 12 has the same N_2 concentration as samples C and E which received a lighter doping, this phenomena is still under investigation.

CONCLUSION

High temperature heat treatments with N_2 doping have proven to improve performance of SRF Nb cavities. The Fermilab furnaces that are used for N_2 doping have been described with a major focus on how the N_2 doping is executed automatically, and how each step is implemented by the furnace PLC control.

Typical plots of a N_2 doping recipe have been presented and discussed. Finally surface analyses with SIMS are also shown to prove the successful introduction of N_2 in the Nb (Nb nitrides on the first μm and interstitial N_2 in the bulk).

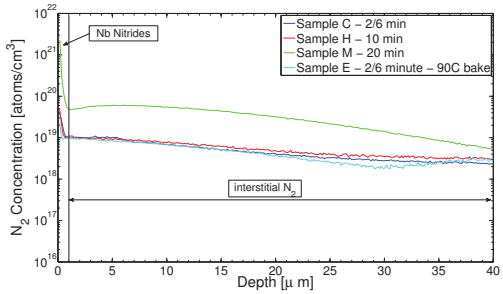

Figure 12: N_2 concentration in Nb samples baked with different recipes.

Big TM will be upgraded to a higher operating temperature, up to 1300C. Future work with the furnace may involve developing new recipes possibly involving new doping gases and optimizing the N_2 doping recipe for 650 MHz single and 5 cells.

ACKNOWLEDGMENT

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