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STATUS OF HIGH TEMPERATURE VACUUM HEAT TREATMENT PROGRAM AT IPN ORSAY

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Abstract

In the framework of ESS, a vacuum furnace dedicated to High Temperature Heat Treatment under Vacuum (VH2T2) of SRF bulk Nb cavities was developed and commissioned in May 2016. This furnace is currently used for interstitial hydrogen removal (10h00 @ 650 °C) of two type of cavities: 1) the whole series of 26 ESS 352 MHz spoke resonators equipped with their Ti LHe tank well, 2) some prototypes of ESS high beta and medium beta cavities. Up to know IPN Orsay VH2T2 (10h00 @ 650 °C) was successfully applied to more than 16 cavities. In this paper we will first report about these VH2T2 tests. Finally, we have just started testing nitrogen infusion and nitrogen doping processes on samples and 1.3 GHz cavities: the preliminary results will be discussed.

INTRODUCTION

SRF cavities made of high purity bulk niobium (e.g. Nb with Residual Resistivity Ratio RRR >200) showed in the early 1990 a decrease by a factor ~10-30 of the unloaded quality factor Q_0 when cooled down to cryogenic temperatures under certain conditions. This so-called Q_0-disease was observed in many laboratories [1-4] for various cavity shapes resonating at frequencies in the range 500 MHz-5.6 GHz. According to this early study [1], Q_0-disease is observed when two conditions are fulfilled during the cool down of the cavity: 1) slow cooling rate $\Delta T/dt << 1$ K/min., 2) dwell time t_D or cryogenic cool down duration from 170 K to 80 K higher than 1h00. Furthermore, no more Q_0 decrease is observed for t_D > 3h00: the phenomenon is characterized by a saturation effect. This Q_0-disease is attributed to the formation of $NbH_{0.7}$ hydride precipitate [1, 5] on the cavity RF surface. More precisely, the solubility limit of interstitial hydrogen in niobium is $C_{Hmax1}=4.10^4$ at. ppm at T_1=300K and decreases drastically to C_{Hmax2}=5 at. ppm at T_2=100K. Beyond C_{Hmax}, $NbH_{0.7}$, hydride precipitation [5] was experimentally observed (i.e. 100-400 at. ppm) on SRF 1.3 GHz Nb cavities [1] after a Buffered Chemical Polishing (BCP). More recently, using a cryogenic confocal laser scanning microscopy, Barkov [6] studied in-situ the kinetics of $NbH_{0.7}$ formation on a RRR=300 Nb with C_H= 2323 at. ppm: $NbH_{0.7}$ precipitates ~5-10 µm thick and lateral dimensions ranging from 10 µm to 50µm, were observed at T=140 K. Finally, High Temperature (e.g. θ>500 °C) Heat Treatment under Vacuum (VH2T2) of SRF bulk Nb cavities [1] reduces the interstitial hydrogen concentration C_H to less than the threshold C_{Hmax}=5 at. ppm, resulting in a cure of the resonator from Q_0-disease.

FURNACE COMMISSIONING AND QUALIFICATION OF THE PROCESS

The furnace was developed in the framework of ESS [6]: it is dedicated to VH2T2 of ESS spoke [7] and elliptical cavities. The goal of VH2T2 is twofold: 1) reduce C_H in Nb below C_{Hmax2}, 2) relieve the residual stresses due to cold work and plastic deformation of Nb during the cavity fabrication in order to restore initial RRR value of Nb sheets. The main objectives of the program are :1) master the techniques of VH2T2, 2) optimize the process parameters, including post-heat treatment of cavities equipped with titanium made Liquid Helium (LHe) tank , 3) improve the reliability of VH2T2 process for large scale, 4) progress in understanding of the physical phenomena underlying Nitrogen Doping (N-Doping) and Nitrogen Infusion (N-Infusion) processes, 5) Qualify and master N-Doping and N-Infusion for reliable application to SRF cavities at large scale.

Furnace Description and Commissioning

More than 26 parameters where specified for the furnace shown in Fig. 1. Due to space limitation, only the main parameters of the furnace are listed in Table 1, which includes also the values achieved during the commissioning at IPNO in May 2016 [8].

Table 1: Main Performance Specifications and Commissioning Results

Parameter	Specification	Achieved
Temperature θ (°C)	20-1400	20-1400
Temp. uniformity (°C)	+/-5	+/-3
Heating rate (°C/min.)	1-10	1-20
Residual pressure (mbar) at θ>600 °C	5.10^{-7}-10^{-6}	5.10^{-7}-10^{-6}
Pumping speed of hydrogen (l/s)	1410^3	n.a
Volume of the thermal chamber (m^3)	4.5	4.5
Maximum cavity outer diameter (mm)	700	700
Maximum cavity length (mm)	1600	1600

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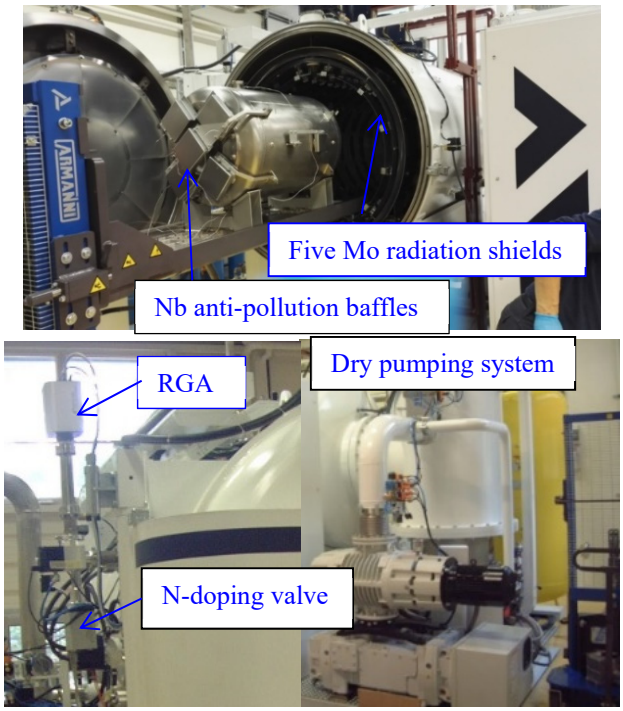

Figure 1: Loading a prototype ESS double-spoke cavity in the furnace.

The pumping system is oil-free (dry) to avoid pollution due to cracking of hydrocarbon and or organic chemicals. It consists of a cryogenic pump (14000l/s – Hydrogen), a roots pump (2050 std m^3/h) and a screw pump (650 std m^3/h). A more detailed description of the furnace and the commissioning results were previousely reported [8]. Briefly, during commissioning all the measured parameters of the intrinsic performances (i.e. residual pressure, thermal performance) are better than the specified.

Qualification Tests With Samples

According to previous studies [9-15], VH2T2 modify strongly Nb material mechanical and thermal properties (e.g. stress versus strain characteristics, yield strength, tensile strength, residual stresses, thermal conductivity) and physical properties (grain size, structural defects). Consequently, SRF cavity mechanical behavior and functional properties (e.g. resonant frequency, field flatness, tuning sensitivity, and RF performances) are impacted. Prior to applying IPNO VH2T2 (See next section for details) to SRF cavities, various qualification tests were performed on several samples: RRR, SIMS analysis, measurements of mechanical and thermal properties at cryogenic temperature. For these tests, we have compared the properties of as received and VH2T2 samples. During the qualification tests and the first VH2T2 of cavities, we systematically used control samples for RRR measurements and SIMS analysis. The qualification tests results were previously [8] discussed in details. We will summarize these results in the following.

Measurements of niobium RRR RRR of niobium was measured using the standard four probes method with reversing the sensing current to eliminate parasitic thermoelectric voltages. The 3.6 mm thick RRR samples have respectively a length and width of 60 mm and 5mm. More than 25 samples from two different suppliers were. It should be stressed that for all these RRR tests, the same reference Nb sample was systematically tested simultaneously with the current batch of samples at each run: the measured RRR values of the reference sample ranged from 400 to 410 leading to an uncertainty of 1.2%. The RRR histogram before and after VH2T2 of 4 samples is illustrated in Fig. 2. These data show that annealing RRR=300-400 niobium cavities at 650 °C for 10h00 reduces slightly material RRR but this RRR reduction is much smaller than that observed in other laboratories where VH2T2 is performed at 800 °C during 3h00 [16].

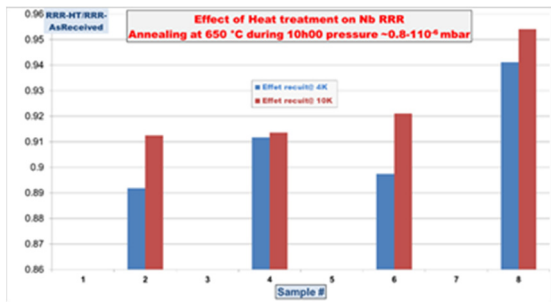

Figure 2: Effect of VH2T2 (10h00 at 650 °C) on Nb RRR.

The degradation of Nb RRR during annealing can be attributed mainly to the oxygen diffusion. During heat treatment of Nb, several phenomena (diffusion, chemical reactions) impacting the oxygen concentration distribution in Nb are observed [17-18]. Ciovati and co-workers [18] measured the profile of oxygen concentration C_O in Nb during VH2T2 at 800 °C: C_O decreases exponentially from ~5.10^{20} atoms/cm^3 on the surface (characteristic length: 50 nm-350nm) to ~5.10^{18} atoms/cm^3. Niobium oxides on the surface are one possible uncontrollable oxygen source. Moreover, at 650 °C, the diffusivity of oxygen in Nb is D_0= 6.410^{-13} m^2/s: this value is ~100 time higher that the diffusivities of N and C in Nb. Consequently, the diffusion lengths of both C and N in Nb are ~10 smaller than that of O. Finally, the sensitivities of Nb electrical resistivity ρ to O, N and C impurities are close (e.g ~4.5 10^{-12} Ω.m/at.ppm). Using the impurities concentration given by the Nb supplier and the expression of Nb electrical resistivity ρ_{10} at T= 10 K as function of impurities (O, N,C) content in Nb and the measured ρ_{10} after HT at 650 °C, we have calculated the oxygen concentration (C_O) to fit the data. The results presented in Table 2, clearly show an increase of C_O by a factor ~10. Note that an oxygen content of 45µg/g corresponds to a concentration C_O= 1.4510^{19} atoms/cm^3 and this value is in the range of the data reported by Ciovati [18].

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Table 2: Effect of Annealing (10h00 at 650 °C) on RRR and Nb Electrical Resistivity at 10 K

Sample	RRR10K Before VH2T2	RRR10K After VH2T2	ρ_{10} (Ω.m) Before VH2T2	ρ_{10} (Ω.m) After VH2T2
4287-2	320	292	4.5010^{-10}	4.9310^{-10}
4288-1	300	275	4.8010^{-10}	5.2410^{-10}
4289-2	342	315	4.2110^{-10}	4.5710^{-10}
4290-2	305	291	4.7210^{-10}	4.9510^{-10}

Pollution Studies We have investigated a potential pollution of the cavity RF surface during VH2T2. We performed SIMS analysis on samples located inside the cavity beam tubes in 2 configurations: for the sample #1 (respectively sample #2) the cavity beam tube is equipped (respectively not equipped) with Nb protection baffles. The data discussed elsewhere [8] showed a pollution of Nb surface with Ti during VH2T2 in the two fore mentioned configurations: 1) for the unshielded sample#1 Ti concentration decreases with the depth (characteristic length ~3-4 µm), 2) for the shielded sample #2, only the sub-surface at a depth ~4-10 nm is polluted with Ti, Fe and Cu. From these data we concluded that: 1) Protection baffles are not necessary even for spoke cavities equipped with their Ti LHe tank, 2) a light BCP (removal ~10-20 µm of the polluted layer) should be used for the cavity post-heat treatment. These conclusions were well confirmed by several spoke cavities cryogenic RF tests.

CAVITY TESTS RESULTS

ESS Double-Spoke Cavity Design

The main RF parameters of the optimized prototype ESS double-spoke [19] cavity shown in Fig. 3 are listed in Table 3.

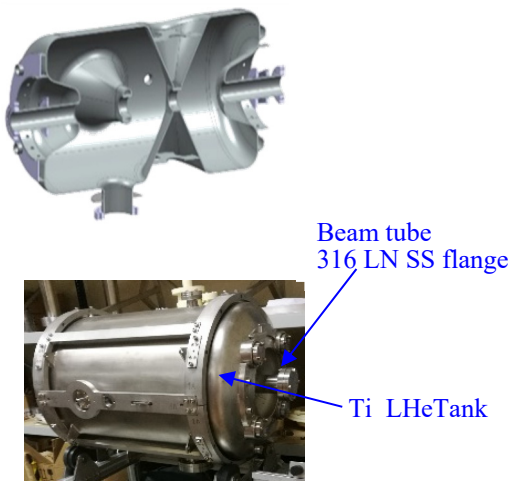

Figure 3: Drawing (A) and photograph (B) of a prototype Double-Spoke cavity for ESS.

Table 3: RF Parameters of ESS Double-Spoke Cavity

Quantity	Symbol (Unit)	Value
Fundamental mode frequency	f (MHz)	352
Accelerating field	Eacc (MV/m)	9
Ratio of Peak surface field to accelerating field	Epk/Eacc	4.3
Ratio of peak surface magnetic induction to accelerating field	Bpk/Eacc (mT/MV/m)	6.9
Geometric factor (fundamental mode)	G (Ω)	130

Results With Cavities

After qualification tests with samples we have applied the VH2T2 (10h00 @ 650 °C) and successfully tested [8] two 1.3 GHz elliptical cavities from CEA, two ESS prototype Double spoke cavities and one MYRTHE single spoke cavity. These previous experimental data confirm the effectiveness of the process for hydrogen outgassing resulting in an improvement of RF performances all the cavities and their final cure against Q_0-diseases. We have then entered the production phase using VH2T2 process for cavities equipped with stainless steel flanges brazed on beam tubes an equipped with their titanium tank. This process will be used routinely for the removal of for interstitial hydrogen of all the 29 serial 352 MHz spoke ESS cavities: until now we have testes 3 prototypes et 7 serial out of 29 resonators ordered (16 already delivered). IPNO standard thermal cycle and post-treatment for spoke cavities with LHe Tank includes 7 steps: 1) Heating up to 300°C @ 5°C/min - Hold @ 300 °C - dwell time : 1h00, 2) Heating up to 650 °C @ 5°C/min - Annealing @ 650 °C, dwell time 10h00, 3) Radiative cooling under vacuum down to 40 °C, 4) Pressurize thermal chamber with Ar up to 900 mbar then to 1013 mbar (filtered air), 5) Cool down to 20 °C, 6) BCP 10 µm, 7) High Pressure Water Rinsing (HPWR), 8) Drying in clean room, 9) Assembly and cryogenic vertical test. Twenty successful hydrogen outgassing of various cavities (prototypes: ESS Double-spoke cavities, MYRTHE single spoke, 1.3 GHz elliptical cavities, serial ESS double spoke cavities, high beta and medium elliptical cavities): the success rate is 100 % for all of these tests. A typical tests results obtained according to the above described process are presented in Fig. 4 for 3 serial double-spoke cavities (DSKP03, DSKP 04 and DSKP06) of ESS and single spoke prototype cavities of MYRRHA (Virginia and Amelia). For both type of spoke cavities with the Ti LHe tank the recorded RF performance are well beyond the specifications with a large margin in terms of unloaded quality factors and maximum achieved accelerating fields.

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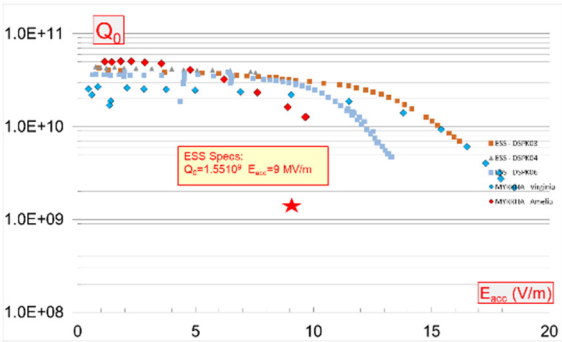

Figure 4: RF performance of ESS and MYRRHA Spoke cavities.

The RF characteristics after hydrogen outgassing at IPNO of 4 serial high beta elliptical cavities of ESS developed by CEA Saclay (HB01, HB02, HB04 and HB05) are shown in Fig. 5. Again the RF performance of these cavities are well beyond the design specifications.

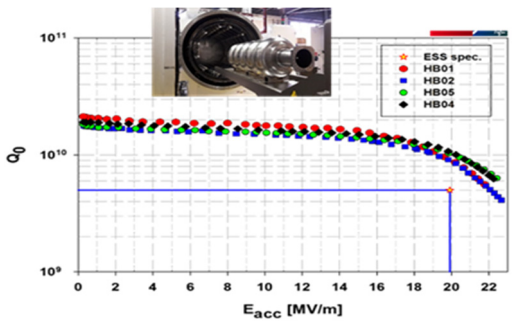

Figure 5: RF characteristics at T=2K of ESS high β elliptical cavities developed by CEA Saclay.

NITROGEN DOPING AND NITROGEN INFUSION

We have just started Nitrogen Doping (N-Doping) and Nitrogen Infusion (N-Infusion) R&D program. This section is devoted to the status of this activity. The main goal of N-Doping or N-Infusion is to improve the RF performance of SRF cavities by reducing the surface resistance R_S resulting in lower RF losses. This surface resistance has two contributions: 1) the so-called BCS surface resistance R_{BCS} which is intrinsic to the material, 2) The residual surface resistance Res which is due, among others parameters, to material structural defects, frozen magnetic flux in the during cool down. N-Doping is based on the strong dependence of the BCS surface resistance with respect to the mean free path l_e of normal electrons: the corresponding curve R_{BCS} vs l_e clearly show of minimum when $l \sim \xi_{BCS}$ where ξ_{BCS} is the intrinsic coherence length of Nb. Accordingly, in order to lower R_{BCS}, one has to reduce l_e down to values close to ξ_{BCS}. The process should be done in clean conditions and in a precisely controlled way in terms of diffusion depth in Nb. The optimum diffusion depth is in the range ~500 nm-1µm. In such conditions R_{BCS} is reduced (i.e reduced RF losses) without decreasing the phonon peak of Nb thermal conductivity (i.e heat transport) otherwise the quench field (due to defects) will be strongly reduced.

N-Doping and N-Infusion in Clean Conditions

These two processes should be performed in clean conditions: 1) clean high vacuum (i.e use dry pumping, see a previous section), 2) used high purity nitrogen and avoid its contamination along the N-injection line. We have used ALPHAGAZ2™ grade Nitrogen. The impurities content of this 99,9999 % purity nitrogen are listed in Table 4.

Table 4: Quality of Nitrogen

Molecule	Content (molar ppm)
H_2O	< 0.5
O_2	< 0.1
$C_n H_m$	< 0.1
CO	< 0.1
CO_2	< 0.1
H_2	< 0.1

Cleaning and Baking of Nitrogen Injection Line

The N-Doping valve- Gas flow @300K could be precisely adjusted in the range: 10^{-10} et 500 mbar.l. s^{-1}. The nitrogen injection line (N-line) made of stainless steel was cleaned and baked according to the following procedure: 1) degreasing with a continuous flow of ethanol, 2) baking with dry pumping during 100 h (temperature: 100 °C-150°C). Then we have qualified the N-line: we measured RGA spectra of the different species in the thermal chamber of the furnace. These tests were performed using the following method: set the furnace pressure at PPN2=10^{-4} mbar and regulate this parameter by means of the micrometric N-Doping valve while the furnace is dynamically pumped (screw pump and roots). The experimental data are illustrated in Fig. 6

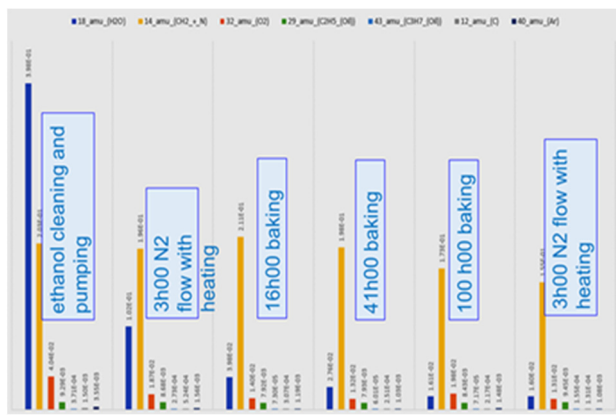

Figure 6: RGA histograms of the different species with reference to nitrogen during the different cleaning steps of the N2-line.

After the cleaning and baking process, water content in Nitrogen (due to contamination from the stainless steel of the nitrogen injection line) was reduces by a factor 25 and the carbon content is negligibly small (~10^{-4}).

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IPNO Nitrogen Infusion Process

The different steps of N-Infusion process of IPN Orsay are: 1) Purge N2 line (0.1 mbar, 30 min.), 2) Cryogenic Pumping, 3) Heating up to 300°C @ 5°C/min - Hold @ 300 °C - dwell time : 0h30, 4) Heating up to 650 °C @ 10°C/min - Annealing @à 650 °C, dwell time 0h30, 5) Heating up to 800 °C @ 5°C/min - Annealing @à 800 °C, dwell time 2h00, 6) Radiative cooling under vacuum down to 150 °C, 7) Temperature regulation @160 °C and Nitrogen Doping @160 °C (0.025 mbar), 8) Cryogenic Pumping, 9) Radiative cooling under vacuum down to 40 °C, 10) Pressurize thermal chamber with Ar up to 900 mbar then to 1013 mbar (filtered air) , 11) Cool down to 20 °C. This process was successfully commissioned including optimization of PPN2 PID regulation.

First Tests of Nitrogen Infusion

We performed 2 N-infusion runs: run#1 with only Nb samples (Fig. 7), run#2 with Nb samples and a 1.3 GHz cavity.

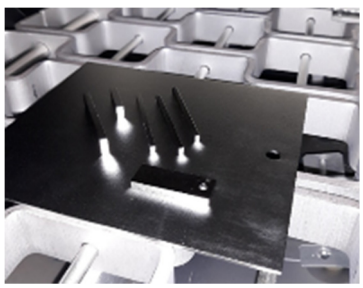

Figure 7: Infusion samples.

The preliminary results (e.g. RGA spectra and SIMS analysis with Nb samples) of these the run#1 did not show any pollution that could potentially impact negatively N-infusion. These results were confirmed for the run #2. Moreover, for the run#2 the confocal microscope micrographs (Fig. 8) did not show any critical contamination of the sample studied.

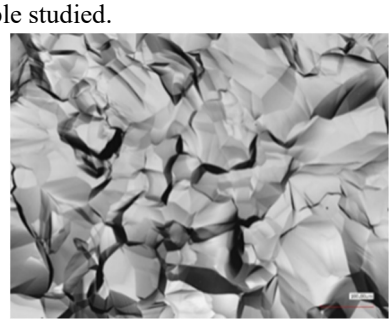

Figure 8: Confocal microscope micrograph.

Finally, the N-infused 1.3GHz cavity was not yet tested: we will report about the corresponding results in the near future.

CONCLUSION

The furnace developed by IPN Orsay in the framework of ESS project was successfully commissioned in May 2016. We have developed hydrogen outgassing process suited for spoke cavities equipped with brazed stainless steel on the beam tubes and titanium LHe tank. This hydrogen outgassing is done at 650 °C for 10h00: we performed successful qualification tests of the process with samples and various cavities (β=1 elliptical resonators and spoke cavities equipped with their Ti LHe tank). We are now in the production phase for ESS. Up to now 20 successful hydrogen outgassing of various cavities (prototypes: ESS Double-spoke cavities, MYRTHE single spoke, 1.3 GHz cavities, Serial ESS double spoke cavities, high beta and medium): the success rate is 100 %. We have just started nitrogen infusion R&D program. Nitrogen infusion process was successfully commissioned. Nitrogen injection line was cleaned, baked and qualified. The preliminary test results (RGA spectra and SIMS analysis with Nb samples) did not show any pollution that could potentially impact negatively N-infusion. First Nitrogen infusion performed with samples and 1.3 GHz cavity: we will report about this first infusion run in the near future.

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