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TUPPO047

POSITRON ANNIHILATION SPECTROSCOPY ON NIOBIUM SAMPLES

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Abstract :

Since the 'baking effect" discovery, a part of the Saclay *R&D* dedicates one's efforts to understand this effect and its correlated question about the High Field Q-slope origin. Experiments on "fast baking" in Oxygen free atmosphere and SIMS analyses have shown that interstitial oxygen diffusion cannot be involved in baking phenomenon.

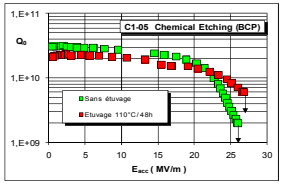
Presence of Niobium vacancy near the surface could contribute to explain such phenomenon. To explore this way, we have performed experiments on niobium samples by means of positron annihilation radiation Doppler broadening spectroscopy. For the first time, an increase of vacancy sites is disclosed at the Nb sample surface after baking. We suggest that the dissociation of vacancy-hydrogen complex is at the origin of the "baking effect". This modification is observed in 100 nm depth under the sample surface, an area where the superconducting RF layer is located.

Context and Summary of Baking Knowledge

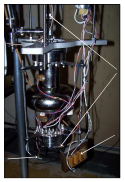
"In-Situ" Baking

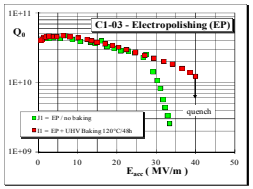
EPAC 1998

Ultra High Vacuum
120 °C / 48 hours




In-Situ Baking in Cryostat with He





- cavity is assembled, ready for RF test
- full equipped with antennas, thermal sensors
- inner part of the cavity is maintained under vacuum
- baking at 120°C for 2 days

"Fast" Baking

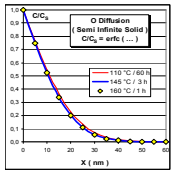
SRF 2005

UHV or Argon (1 atm.)
145 °C / 3 hours
Correspondence with 110°C / 60h
in terms of oxygen diffusion

2nd Fick's law $\frac{\partial C}{\partial t} = D_0 e^{E_A/RT} \frac{\partial^2 C}{\partial x^2}$

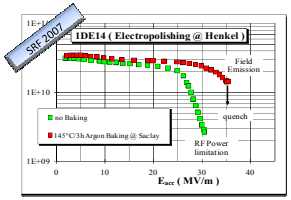
→ analytic solutions — semi infinite solid $C(0,t) = C_S$

$$C(x,t)/C_s = erfc \frac{x}{2\sqrt{D(T)\,t}}$$



- cavity is treated after HPR in Clean Room
- Infra Red Heaters
- cavity under vacuum or filled with Argon (1 atm.)
- baking at 145°C for 3 hours

SRF 2007



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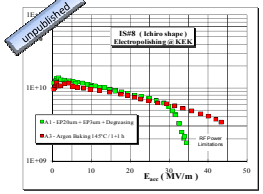
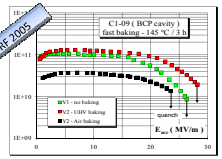


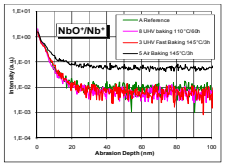
Oxygen Free Atmosphere needed

(Ultra High Vacuum or Argon)

if not : RF degradation after Baking

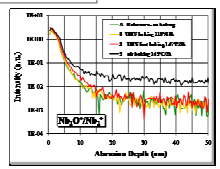
SRF 2005





SIMS analyses on Nb samples

EPAC 2006 — Thin Films 2006



Similar results for plots of NbO^+, Nb_2O^+, Nb_3O^+, Nb_4O^+…

- **No O diffusion @ 10 nm scale – to be avoided**
- **O diffusion @ local scale ? (Nb Vacancy filling)**
- **Vacancy complex dissociation [V.H] ?**
- **Vacancy diffusion (stage III) ?**

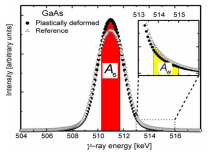
Positron Annihilation Spectroscopy

PAS - DBS (Doppler broadening of positron annihilation radiation)

e^+ → thermalisation (1ps)
→ diffusion
→ trapping in open-volume defects (voids, vacancies, V_{Nb} clusters , dislocation lines, grain boundaries…)
→ annihilation with e^- → 2γ (511 keV) → γ detection

γ Doppler broadening of annihilation line, only due to e^- momentum in propagation direction
(e^+ momentum is negligible due to thermalisation)
sharpness parameter $S = A_S/A_0$ is determined by annihilations with valence electron (low momentum)
wing parameter $W = A_W/A_0$ is determined by annihilations of core electrons (high momentum) and gives chemical information of the annihilation site

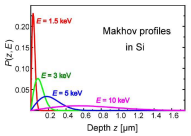
e^+ is easily trapped in **vacancy** (repulsion by lattice ions) increases its lifetime and annihilation is preferentially done with **valence electron** (lack of core electron), smaller momenta, **smaller Doppler shift**, narrow curve, S↑ W↓

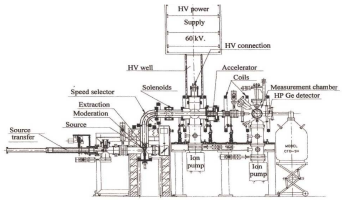


Slow Positron Beam Facility in Orléans

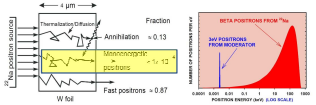
Positron source (^{22}Na)
Moderator (polycrystalline W foil - 4 µm): e^+ (1.10^{-4} @ 3eV)
Extraction (48 eV) – Acceleration (0 to 25 keV)
Beam diameter: 2 mm
γ Detection: high purity Ge detector
Reference sample: UO_2
Vacuum pressure: 5.10^{-8} mbar

positron implantation profiles





e^+ → moderator (W) → 3 eV



Niobium Samples

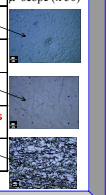
- **single crystal (*SC*)**: beam hole off cut from large grain Nb sheet *provided by X. Singer* – Heraeus RRR 500
- **fine grain (*FG*)**: Tokyo Denkai - RRR 200
  diameter ~ 8 mm
  thickness ~ 1 mm



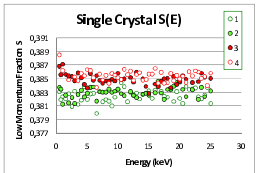


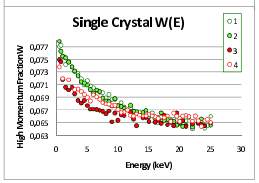
Sample	Niobium	Chemistry	Cutting	Annealing	Chemistry	Baking
1 - 2	Single Crystal Heraeus RRR 500	no	Water Jet abrasive garnet sand (150 µm)	800 °C – 4 hours 1.10^{-7} mbar	BCP - 100 µm	no
3 - 4						145°C / 2 hours Argon – 1 atm
B	Fine Grain Tokyo Denkai RRR 200	EP 500 µm	Water Jet abrasive garnet sand (150 µm)	HF rinse (10% - 30mn) before & after 800 °C – 2 h 30 mn 1.10^{-7} mbar	no	no
G					no	145 °C / 2 hours Argon – 1 atm
C					BCP - 120 µm	no

optical µ–scope (x 50)

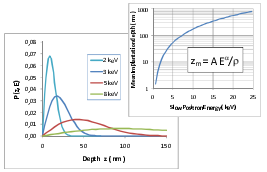


Experimental Results



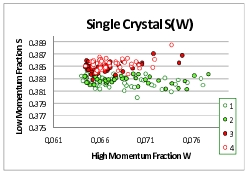


Makhovian profiles in Niobium
(after thermalization & before diffusion)



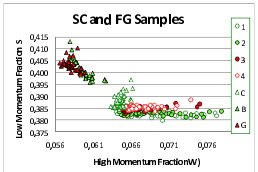
$P(z, E) = 2z/z_0^2 \exp(-(z/z_0)^2$ with $z_0 = z_m/\Gamma(3/2)$

and $A = 2.95$ µg/cm² keV$^{-\alpha}$, $\alpha = 1.7$, $\rho_{Nb} = 8.57$ g/cm³
(A, α) material dependent – Monte Carlo calculation



after baking : S ↑ & W↓ : vacancy number ↑
whole sample but particularly at the RF layer

optical µ–scope picture of fine grain sample C (after 120 µm BCP)



Results for Fine Grain samples:
surface pollution (B and G samples)
more vacancy in C compared to *SC* samples
concentration in excess at interface structures and grain boundaries

Comments on Results

→ **different behaviour at the sample surface between single crystal & fine grain:**
could be explained by a surface pollution for B & G
no hard chemistry after cutting → experiment on fine grain sample C

→ **no vacancy migration**

→ **no vacancy filling** ***(no oxygen diffusion at nm scale)***

→ **vacancy number increases**

Suggestion: ***Hydrogen role - V.H complexes existence - dissociation after baking with hydrogen diffusion (D_0=5.10^{-4} cm²/s → D=2.10^{-5} cm²/s @ 100°C)***
empty vacancies → more traps for positrons

Conclusion

We have experimentally shown that number of vacancies increases after baking treatment on *SC* samples (V_{NB}-H complex dissociation)

Suggested scenario:

- Hydrogen (aqueous species) is absorbed during chemistry; V_{Nb}-H complex creation
- RF losses: Q-drop (like Nb Hydrides in Q-disease)
- Above 100°C complex dissociation and H diffusion out of RF layer → Q-drop removed
- Above 120°C Oxygen diffusion in material → V_{Nb}-O complex → Q-drop restored

- Additional BCP on fine grain samples B & G (to confirm C result)
- Baking of *FG* samples (C,B & G) to confirm baking results on *SC*

- Complementary experiments (ERDA) to follow hydrogen atom