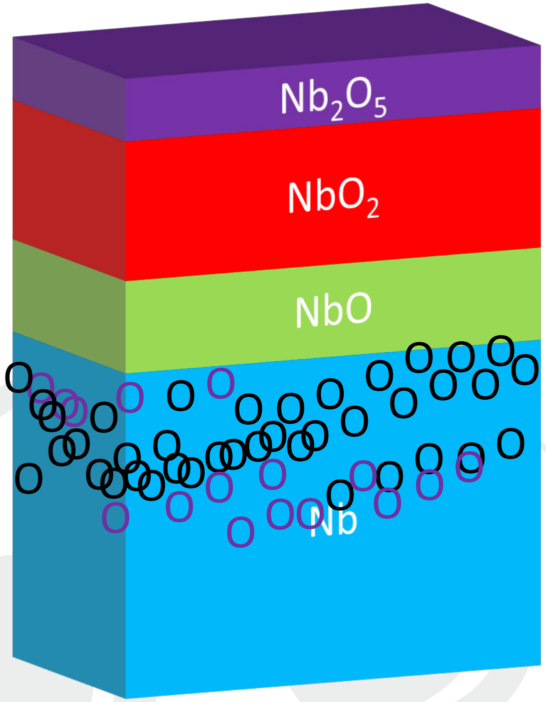
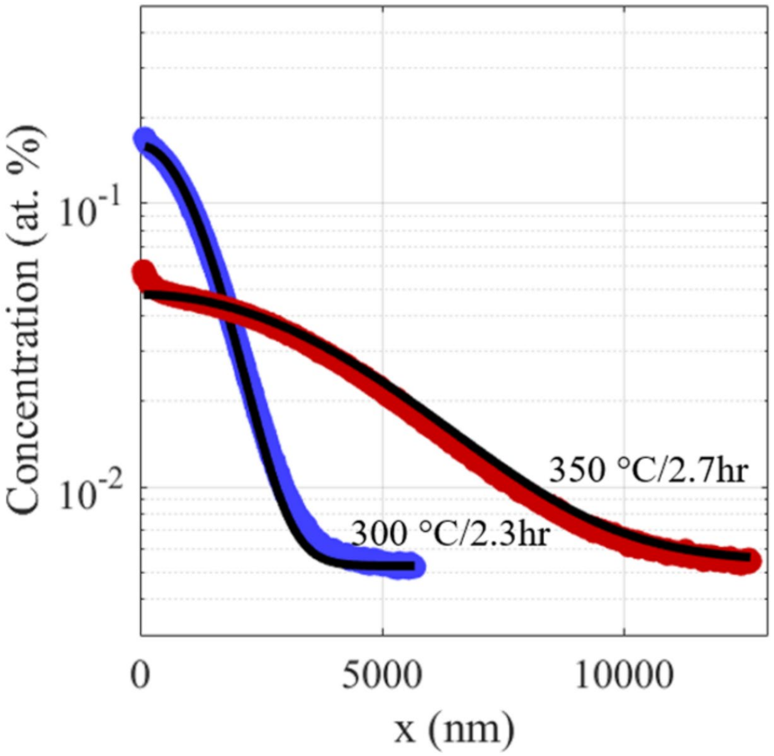
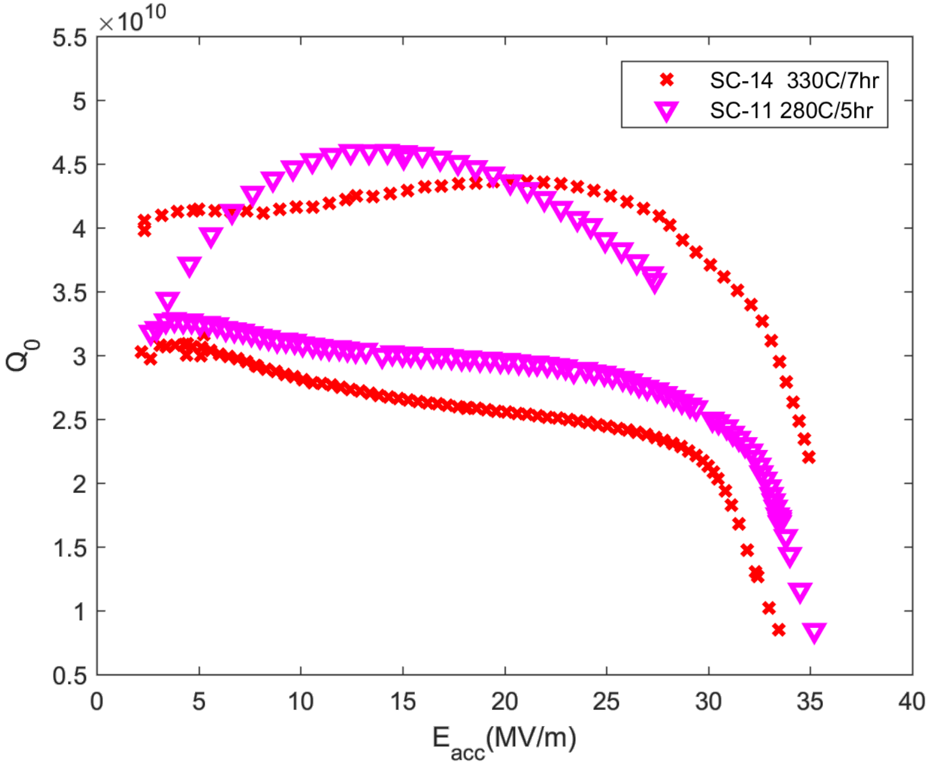
Modeling O Diffusion During Nb Oxide Dissolution







Eric Lechner

Monday, October 10, 2022

Outline

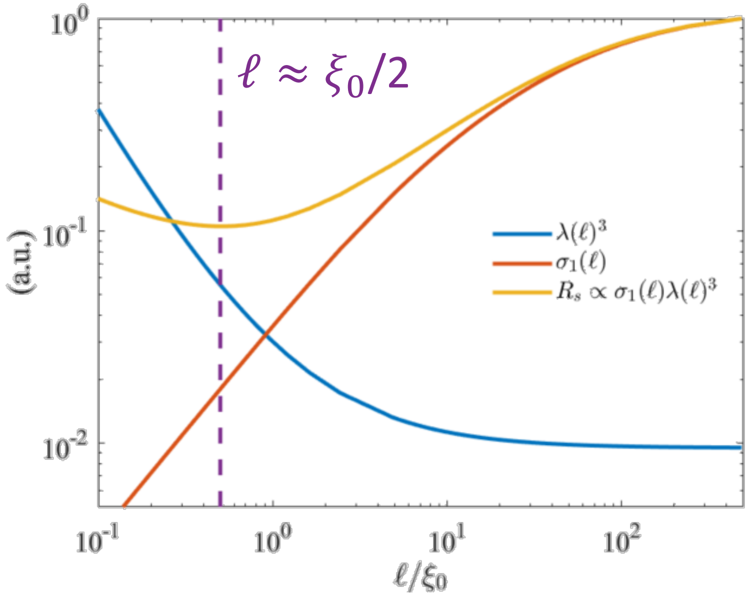
- Motivation
- Modeling O Diffusion
- Where next?

Motivation - Impurity Alloying

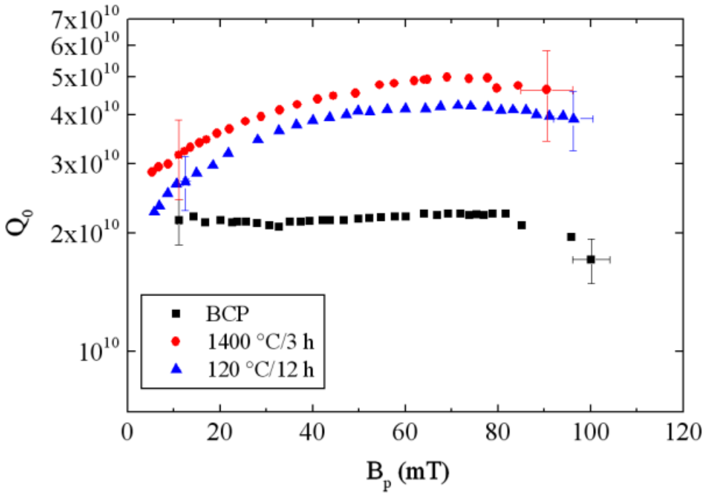
Why introduce impurities?

- 2-4 times more efficient
- Only way to obtain high Q_0

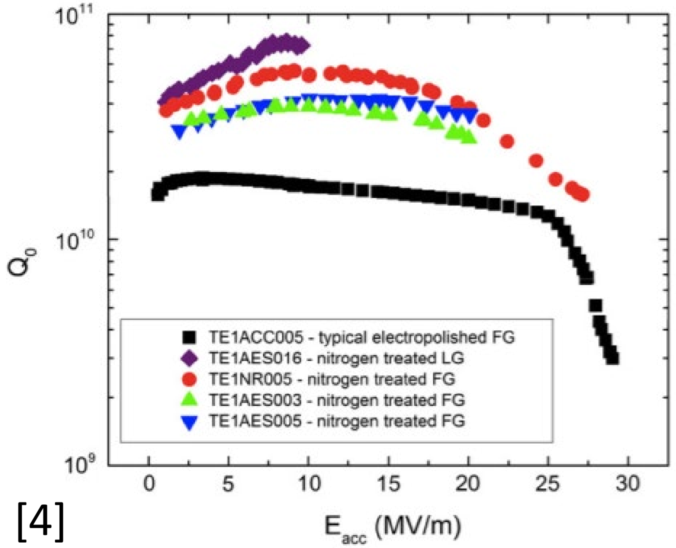
Calculated from [1] $R_s = \frac{1}{2}\mu_0^2\omega^2\lambda^3\sigma_1,$

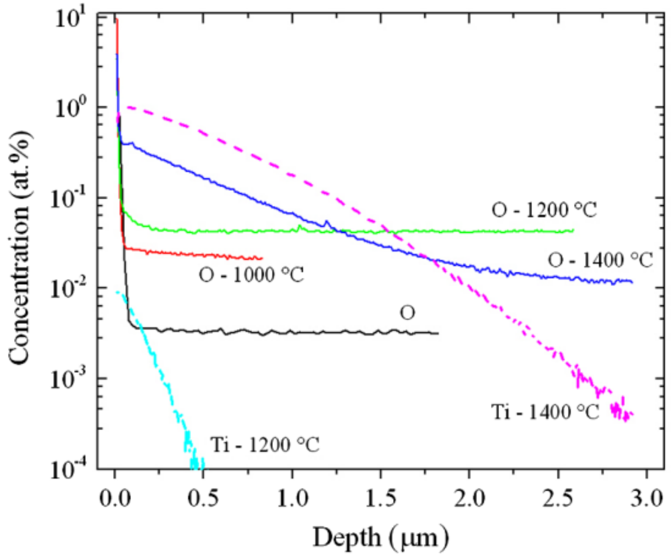


[2] Ti impurities

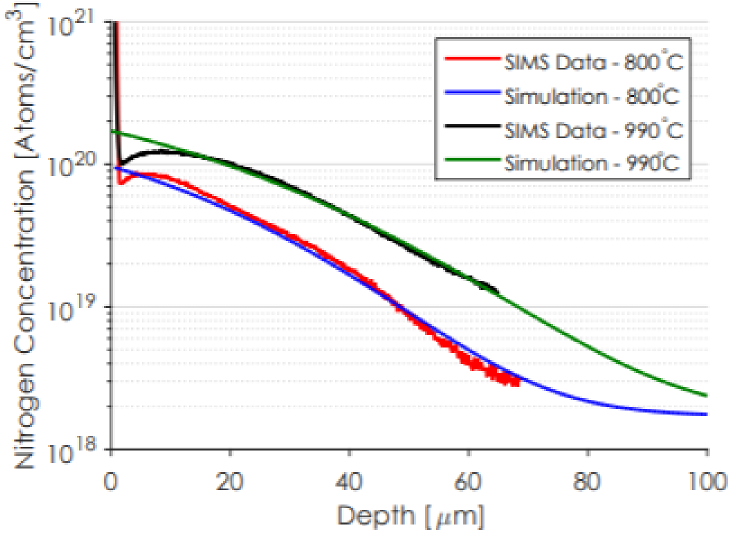


[3] N impurities





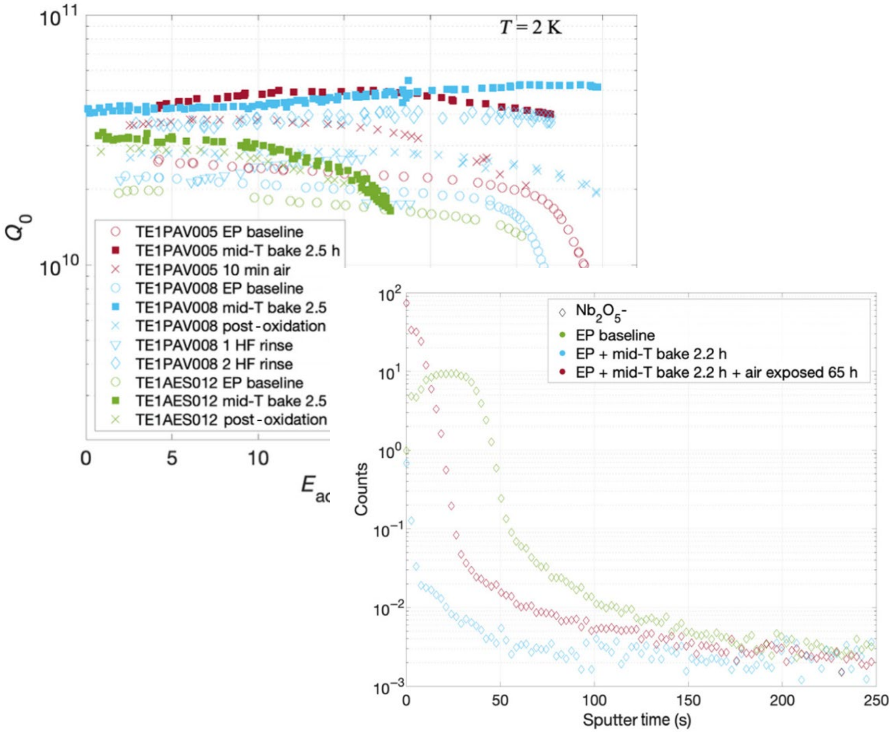
[4]



[1] Kubo, Takayuki. *Physical Review Applied* 17.1 (2022): 014018.
[2] Dhakal, P., et al. *Physical Review Special Topics-Accelerators and Beams* 16.4 (2013): 042001.
[3] A. Grassellino et al. *Supercond. Sci. Technol* 26.10 (2013): 102001.
[4] D. Gonnella, Daniel, et al. *Proceedings of IPAC2016, Busan, Korea* (2016).

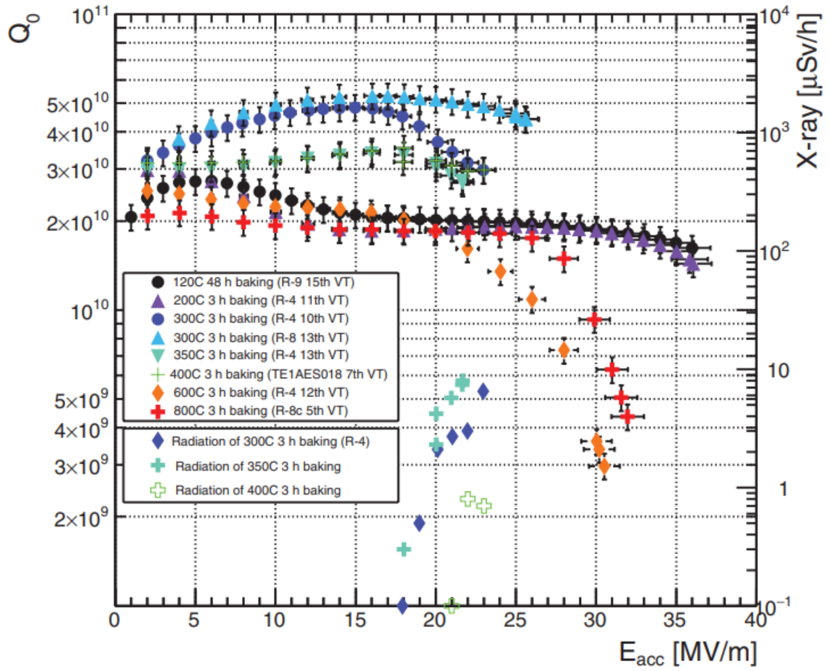
Recent Experiments

[1]



- Efficacy of cavities heat treated at 250-400 C
  - Nb pentoxide dissolution observed

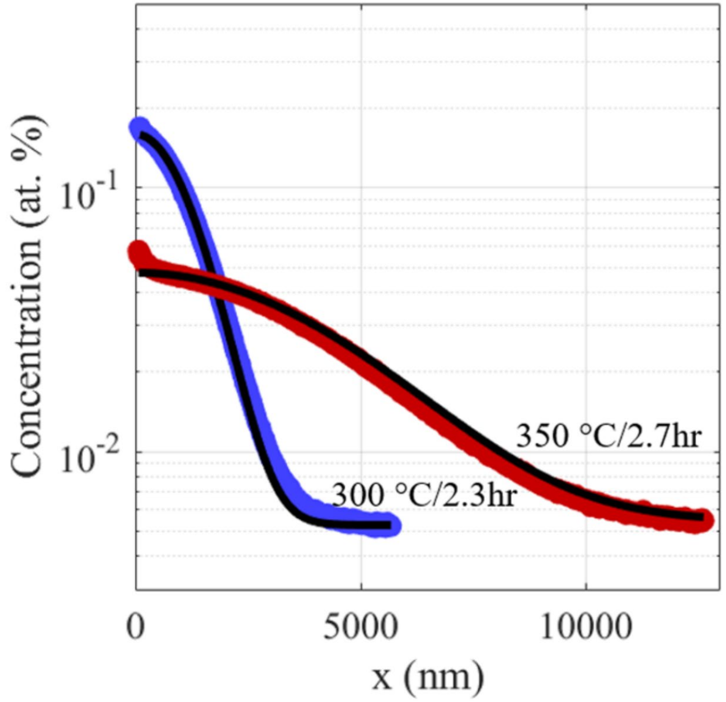
[2]



- Cavities heat treated between 200-400 show an enhancement in Q, but at higher temperatures it resembles a baseline cavity
- Consistent with an alloying agent being introduced and diffused away

[3]

O impurities

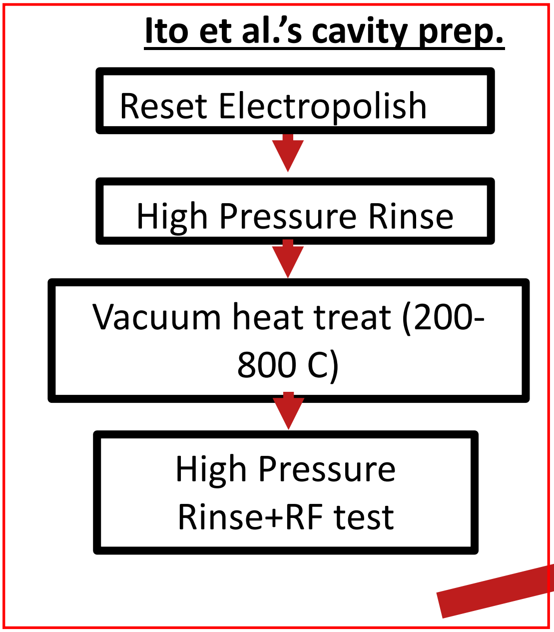


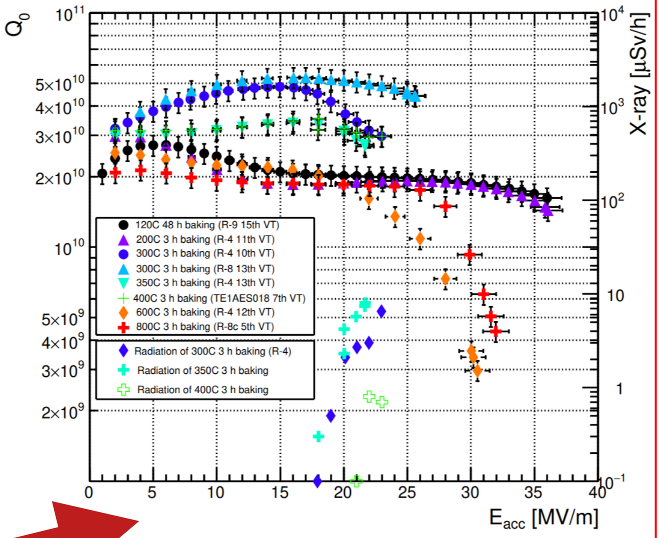
- Oxygen shown as major contributor at ~0.1 at %. A similar concentration of N in N doped cavities.
- Consistent with Ciovati's model of Nb pentoxide dissolution and O diffusion.

[1] Posen, S., et al. *Physical Review Applied* 13.1 (2020): 014024.
[2] Ito, H., et al. *Progress of Theoretical and Experimental Physics* 2021.7 (2021): 071G01.
[3] Lechner, E. M., et al. *Applied Physics Letters* 119.8 (2021): 082601.

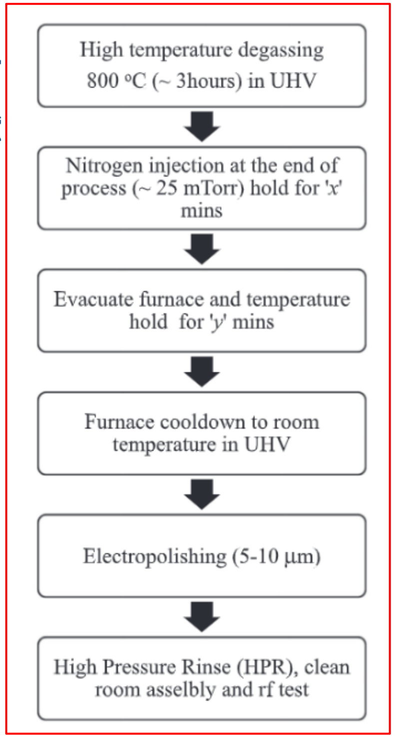
Motivation – Simplified Processing

[1] O-alloying



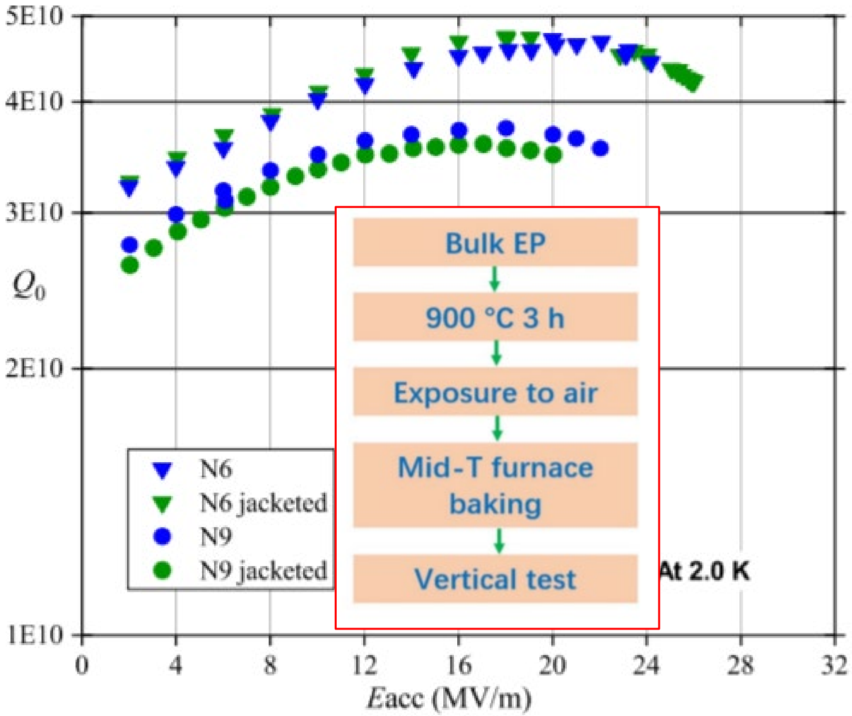


[2] N-doping



COMPLICATED!

[3] How simple can O-alloying get?

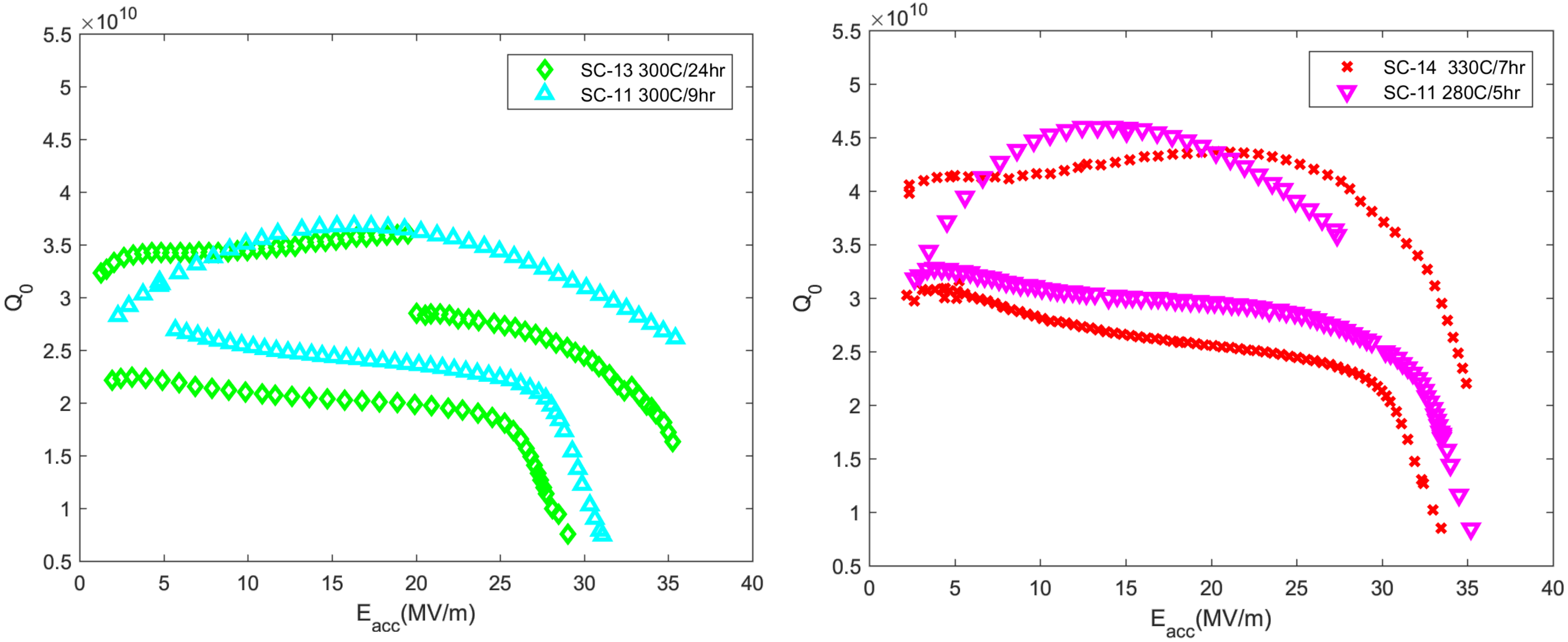


SIMPLE!

[1] H. Ito et al. Progress of Theoretical and Experimental Physics, 2021;, ptab056
[2] P. Dhakal *Physics Open* (2020): 100034.
[3] F. He, et al. *Superconductor Science and Technology* 34.9 (2021): 095005.

High Q_0 and High E_{acc}

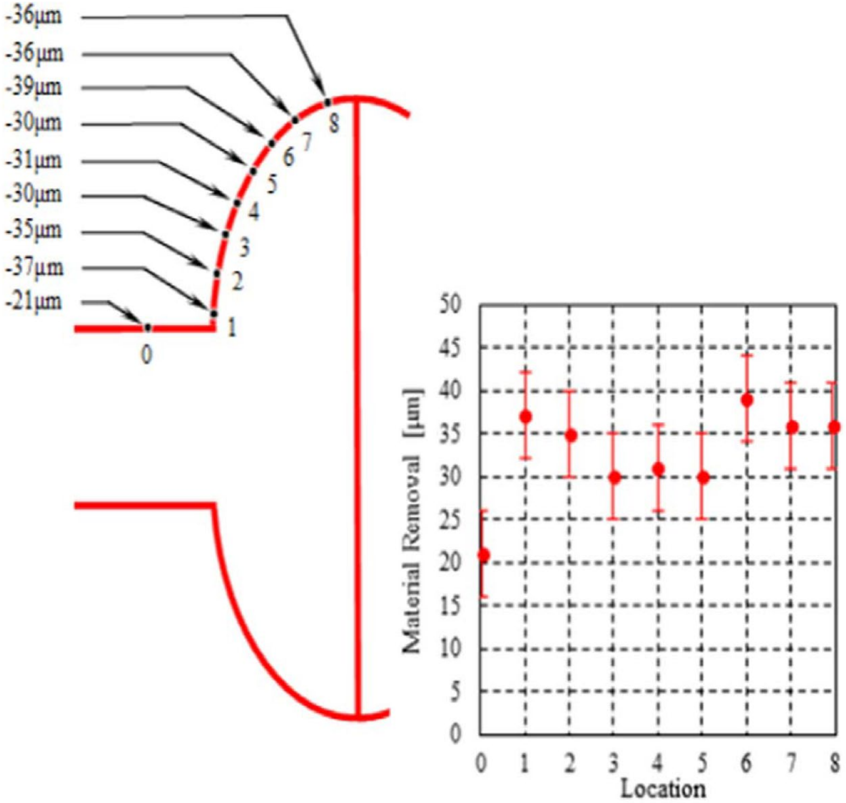
1.3 GHz Single Cell RF Tests



- High Q
- E_{max} that match or exceed the baseline max

Benefits – Conformal

[1]



[2]

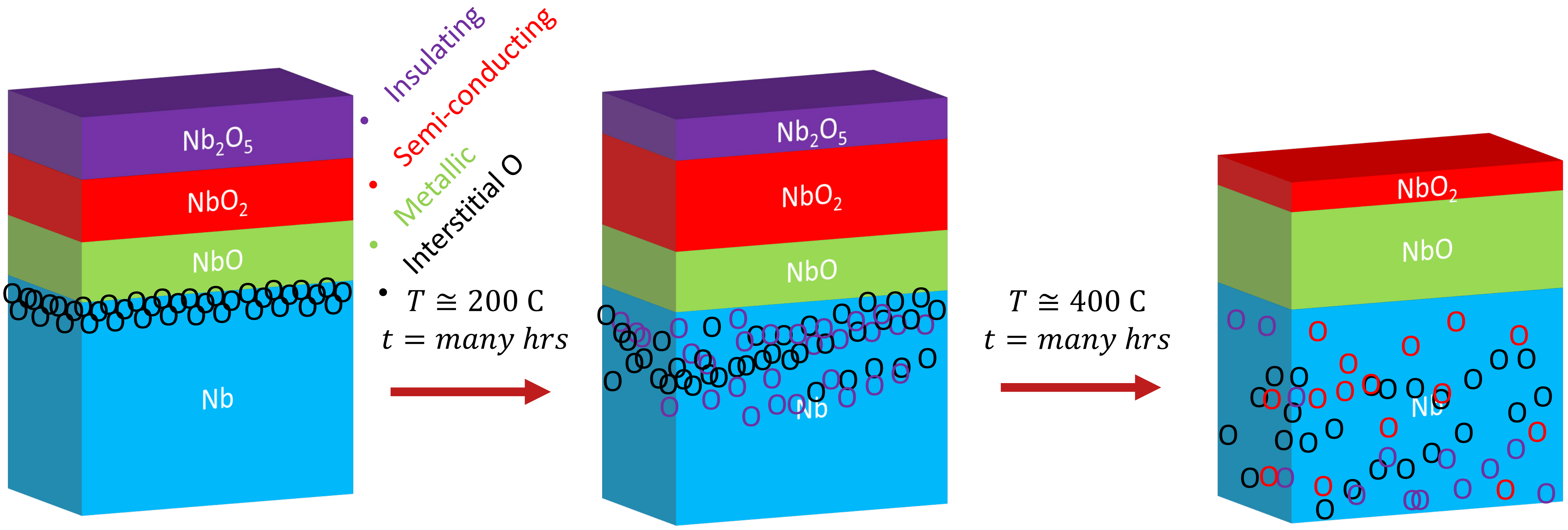
- N-alloying requires an additional electropolishing step to remove lossy nitrides.
- On complicated shapes this may result in non-uniform impurity profiles exposed to the RF surface. No electropolishing required with O-alloying
- The oxygen source is inherently conformal to the cavity shape

[1] Crawford, Anthony C. Nucl. Instrum. Methods. Phys. Res. B 849 (2017): 5-10.
[2] De Silva, S. U., and Jean R. Delayen. *Physical Review Special Topics-Accelerators and Beams* 16.8 (2013): 082001.

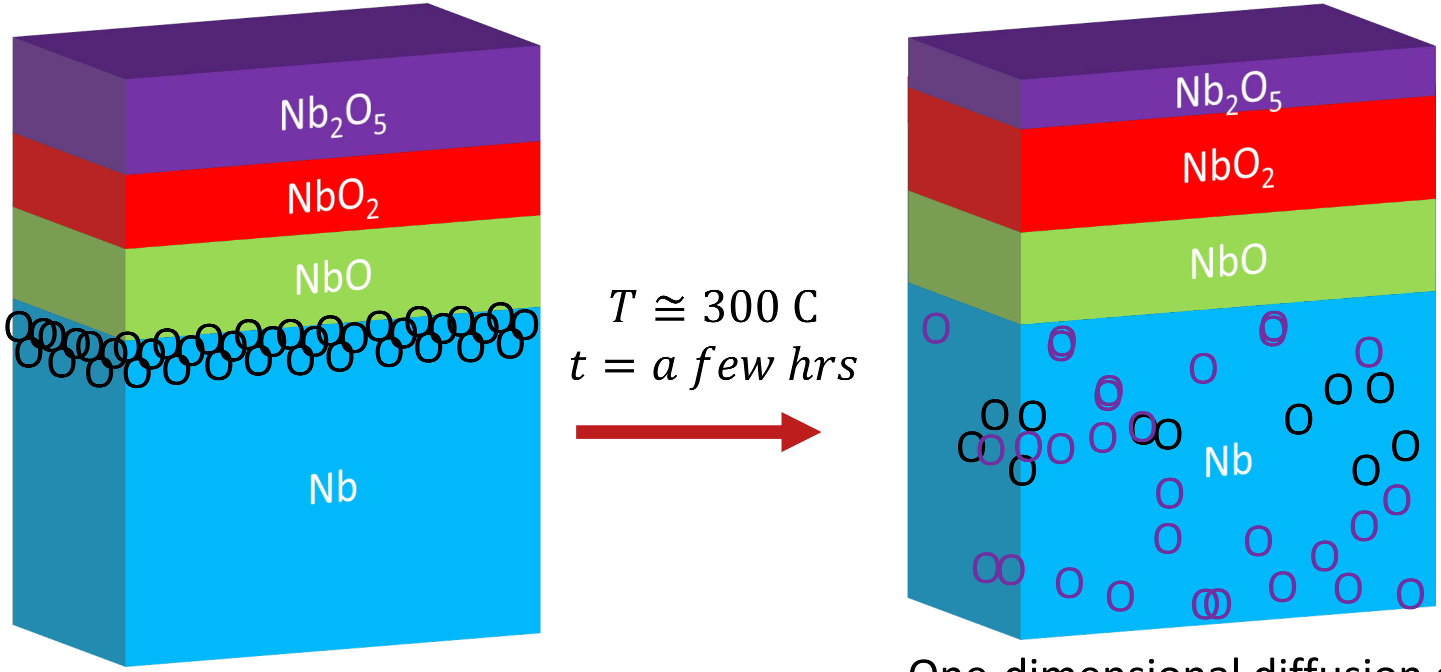
Outline

- Motivation
- **Modeling O Diffusion**
- Where next?

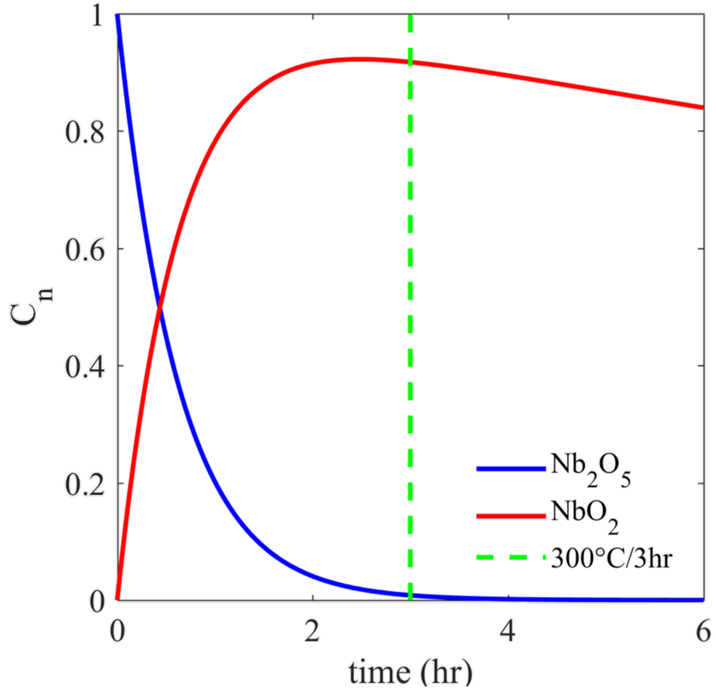
Native Oxide Dissolution



Ciovati's Native Oxide Dissolution and Diffusion Model



Estimate from King [2]



One-dimensional diffusion equation [1]

$$\frac{\partial c(x,t)}{\partial t} = D(T)\frac{\partial^2 c(x,t)}{\partial x^2} + q(x,t,T). \qquad c(x,t) = v(x,t) + u(x,t)$$

Solutions to the diffusion equation

$$v(x,t) = \frac{v_0}{\sqrt{\pi D(T)t}} e^{-x^2/(4D(T)t)} + c_\infty \; - superficial\ dissolved\ oxygen$$

$$u(x,t) = \frac{u_0}{\sqrt{\pi D(T)}} \int_0^t \frac{k(T)e^{-k(T)s}}{\sqrt{t-s}} e^{-x^2/(4D(T)(t-s))} ds \; - oxide\ dissolution$$

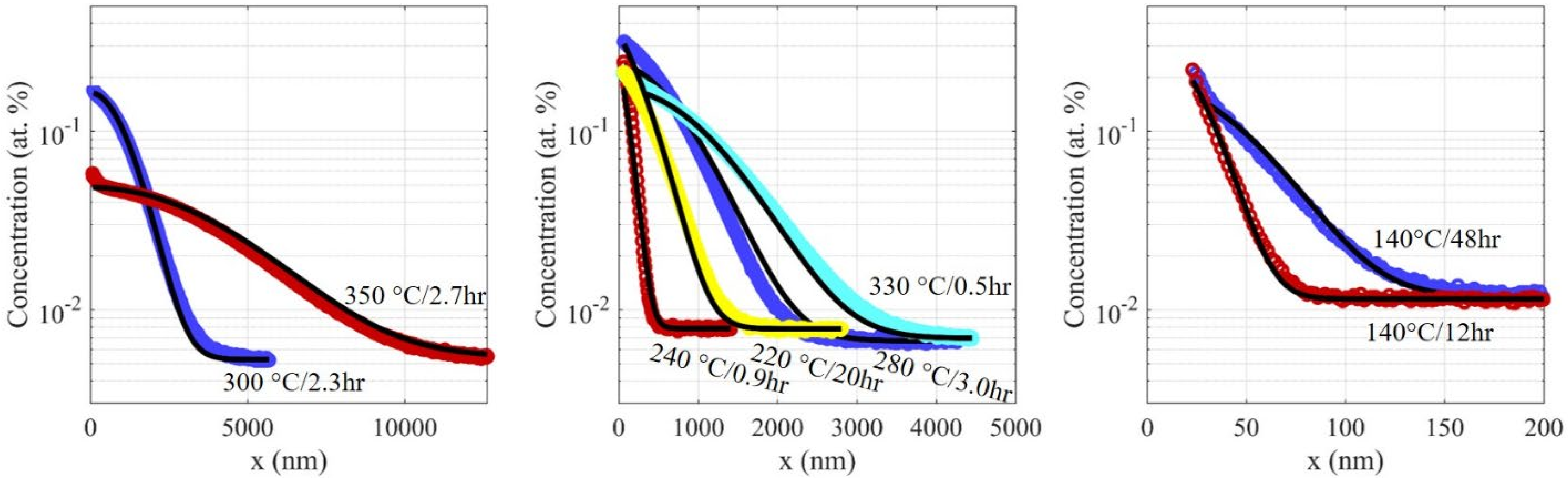
Arrhenius equations

$$D(T) = D_0 e^{-E_{aD}/RT} \; - oxygen\ diffusion$$

$$k(T) = A e^{-E_{ak}/RT} \; - \; oxide\ dissolution$$

[1] Ciovati, Gianluigi. *Applied physics letters* 89.2 (2006): 022507.
[2] King, B. R., et al. *Thin Solid Films* 192.2 (1990): 351-369.

Application of Ciovati's Model – SIMS O Concentration Depth Profiles



- Measurements are consistent with Ciovati's model
- May be able to tune interstitials by changing only temperature and time of vacuum anneal, but missing components of the slower dioxide dissolution.

superficial dissolved oxygen

$v_0 = 3.5$ O % nm

$E_{aD} = 119.9$ kJ/mol

$D_0 = 0.075$ cm^2/s

oxide dissolution

$u_0 = 200$ O % nm

$E_{ak} = 131$ kJ/mol

A = 0.9 ×10^9 1/s

Model of oxide dissolution - Extension

$$-\frac{dA}{dt} = k_1 A; \; -\frac{dB}{dt} = k_2 B - 2k_1 A; \; -\frac{dC}{dt} = -k_2 B$$

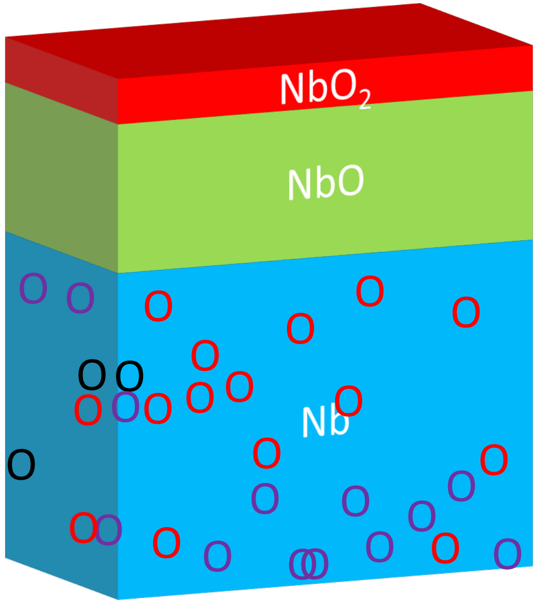
$$Nb_2O_5 \xrightarrow{k_1} 2NbO_2 + O$$

$$2NbO_2 \xrightarrow{k_2} 2NbO + 2O$$

$$A = A_0 \exp(-k_1 t)$$

$$B = \frac{2A_0 k_1 \left(\exp(-k_1 t) - \exp(-k_2 t)\right)}{k_2 - k_1} + B_0 \exp(-k_2 t)$$

$$C = 2A_0 \left(\frac{k_1 \exp(-k_2 t) - k_2 \exp(-k_1 t)}{k_2 - k_1} + 1 \right) + B_0 \left(1 - \exp(-k_2 t)\right) + C_0$$

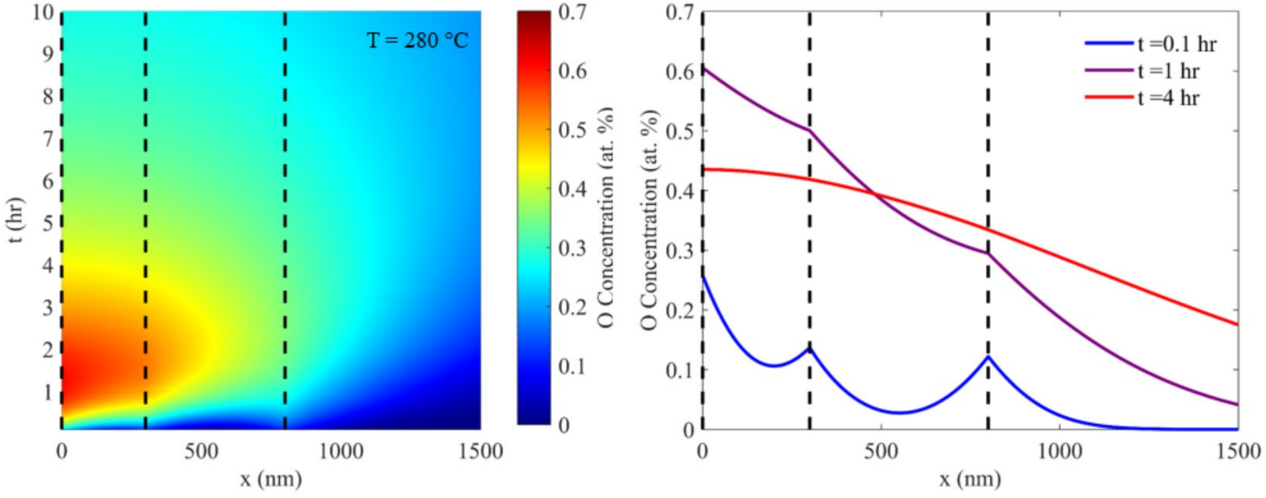


$$\frac{\partial c(x,t)}{\partial t} = D(T) \frac{\partial c(x,t)}{\partial x^2} + \sum q_X(x,t)$$

$$k_2 B \propto q_2(t)$$

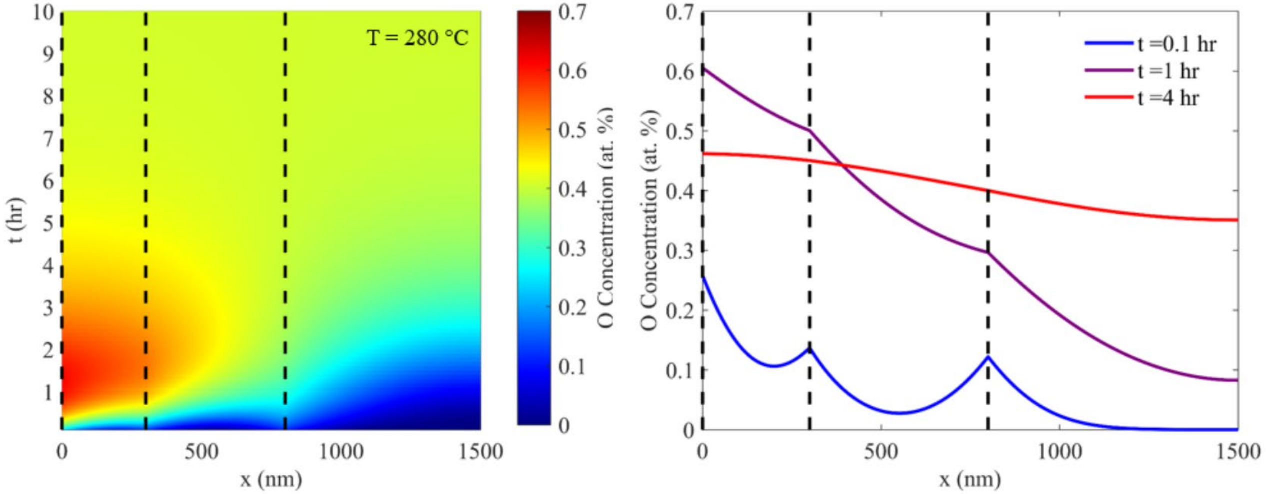
Application to Arbitrary Sources in Semi-infinite Slabs and Films

Semi-infinite Slabs



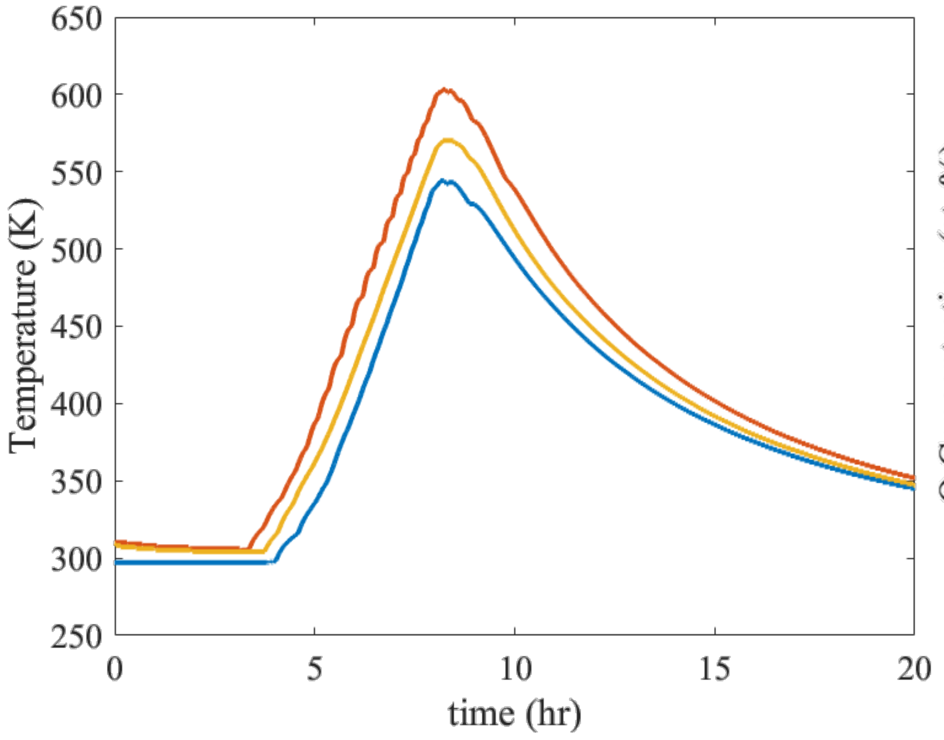
$$
\begin{aligned}
c(x,t) &= \sum \frac{v_0}{\sqrt{(4\pi D t)}}\left(\exp\left(-\frac{(x-a_n)^2}{(4Dt)}\right)\right. \\
&\left. + \exp\left(-\frac{(x+a_n)^2}{(4Dt)}\right)\right) + \int_0^t \frac{\gamma(s)}{\sqrt{(4\pi D(t-s))}}\left(\exp\left(-\frac{(x-a_n)^2}{(4D(t-s))}\right)\right. \\
&\left. + \exp\left(-\frac{(x+a_n)^2}{(4D(t-s))}\right)\right) ds + c_\infty
\end{aligned}
$$

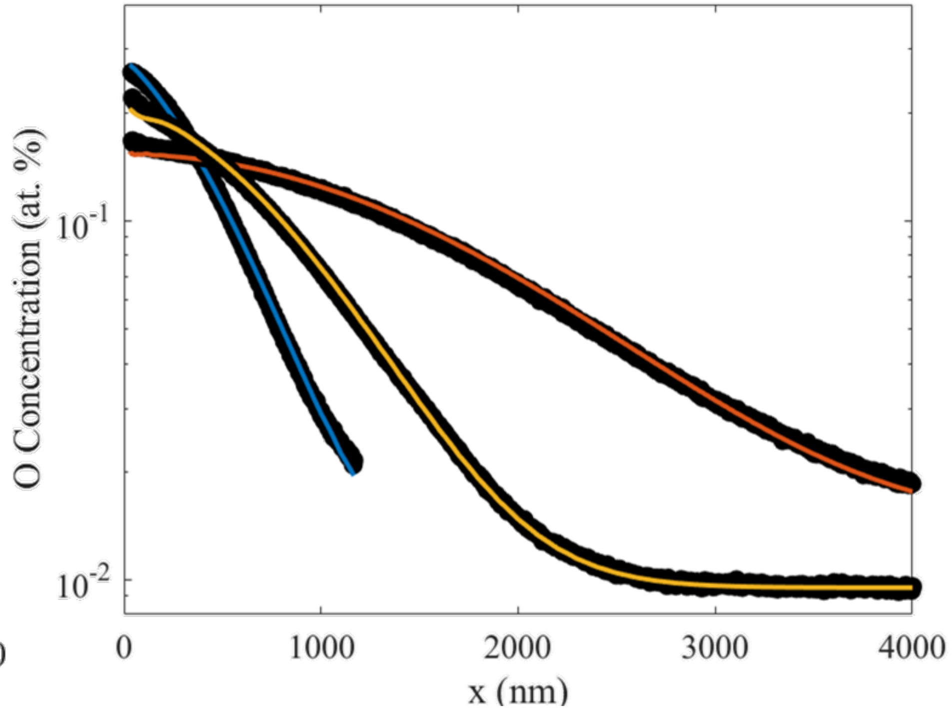
Films with O Diffusion Barrier



$$
\begin{aligned}
c(x,t) &= \sum_{k=-\infty}^{\infty}\sum_{n=1}^{m} \frac{v_0}{\sqrt{(4\pi D t)}}\left(\exp\left(-\frac{(x-a_n-2kd)^2}{(4Dt)}\right)\right. \\
&\left. + \exp\left(-\frac{(x+a_n-2kd)^2}{(4Dt)}\right)\right) + \int_0^t \frac{\gamma(s)}{\sqrt{(4\pi D(t-s))}}\left(\exp\left(-\frac{(x-a_n-2kd)^2}{(4D(t-s))}\right)\right. \\
&\left. + \exp\left(-\frac{(x+a_n-2kd)^2}{(4D(t-s))}\right)\right) ds + c_\infty .
\end{aligned}
$$

Short Heat Treatments





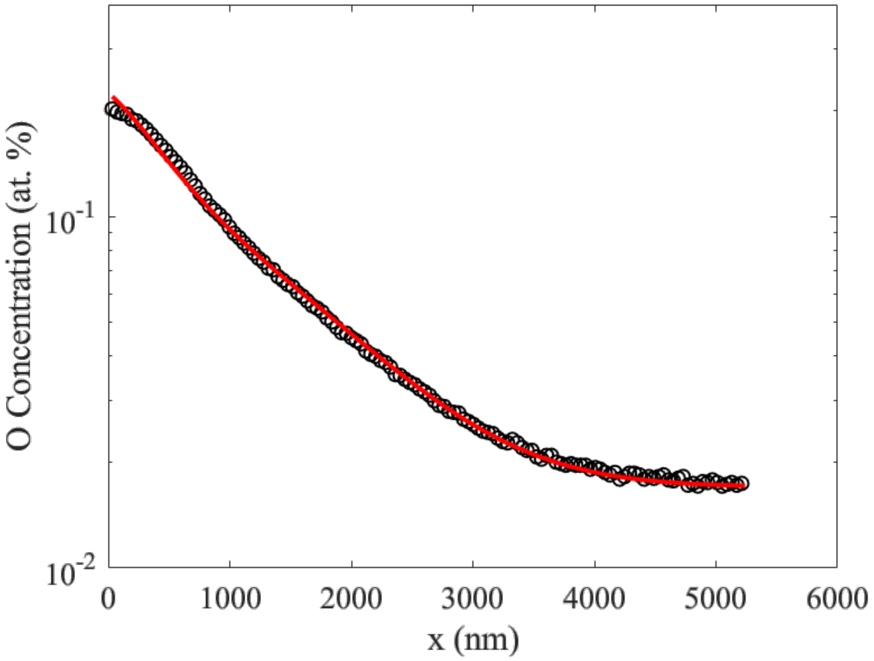
- Suppose you want to employ a short heat treatment at higher temperatures to promote greater oxide dissolution but keep a short diffusion profile. In this case heating and cooling steps may be relevant.

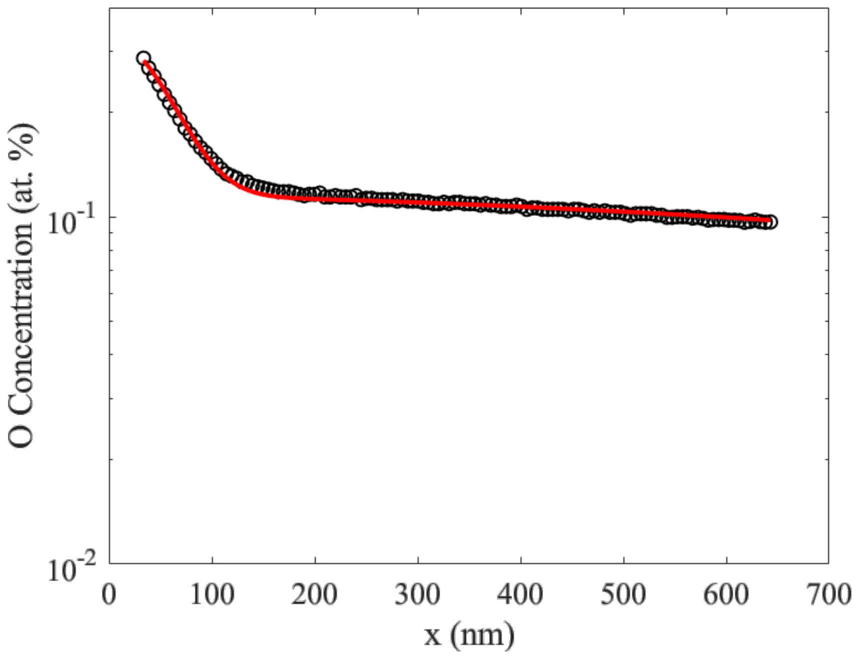
$$\frac{\partial c(x,t)}{\partial t} = D\big(T(t)\big)\frac{\partial c(x,t)}{\partial x^2} + q(t,T(t))$$

$$-\frac{dA}{dt} \propto q = u_0 k\big(T(t)\big)\exp\left(-\int_0^t k\big(T(s)\big)\,ds\right)\delta(x).$$

Jefferson Lab

Two-step Baking





1. Start with the input of Ciovati's model for the first temperature and time
2. Re-oxidize the surface
3. Replenish the sources in the model and advance the solution numerically.

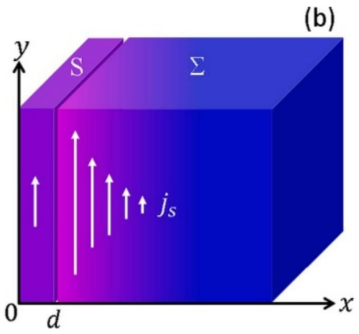
$$\frac{\partial c(x,t)}{\partial t} = D(T)\frac{\partial c(x,t)}{\partial x^2} + q(t,T)$$

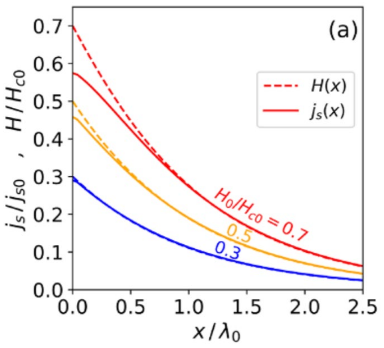
Outline

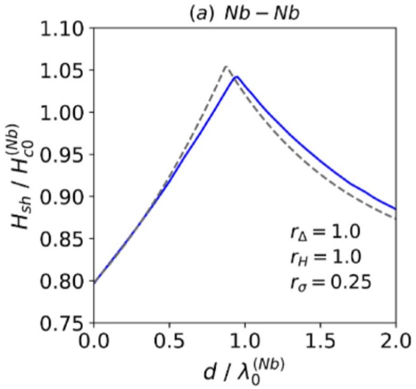
- Motivation
- Modeling O Diffusion
- **Where next?**

Where Next? – Shallow Impurity Profiles

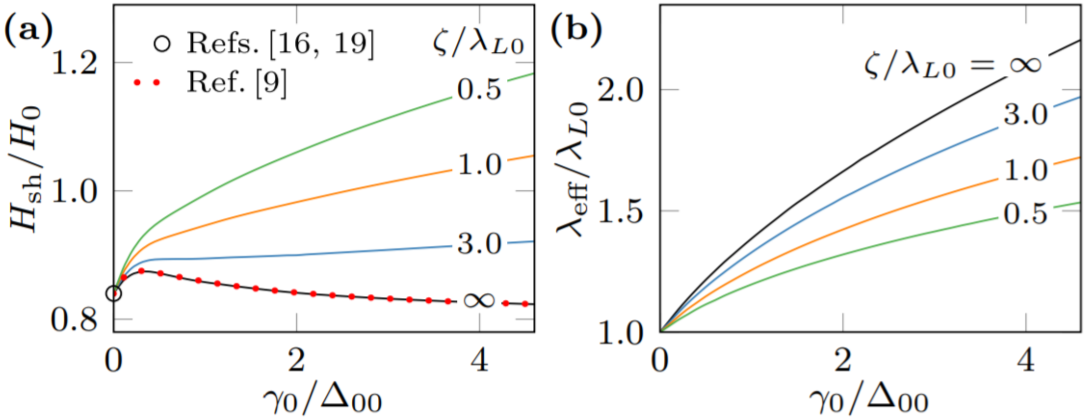
[1] S-S' "Dirty" Layer Model



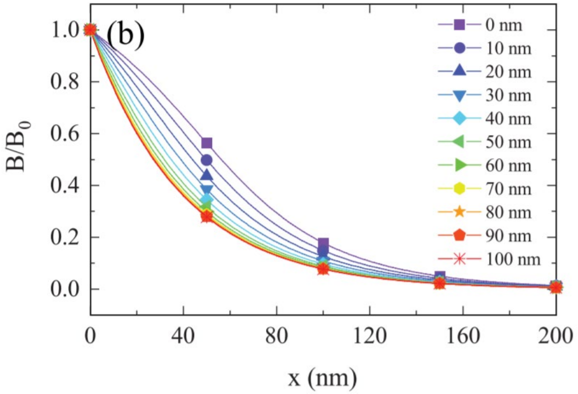


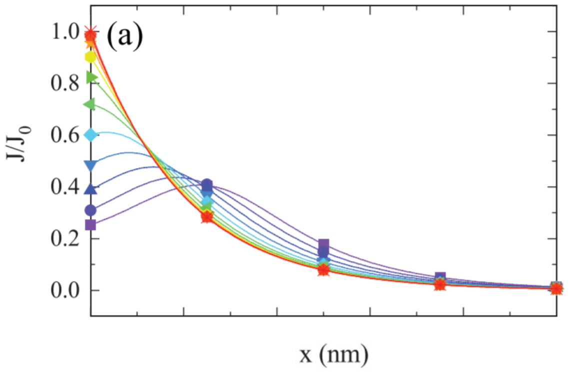


[3] Impurity profile model

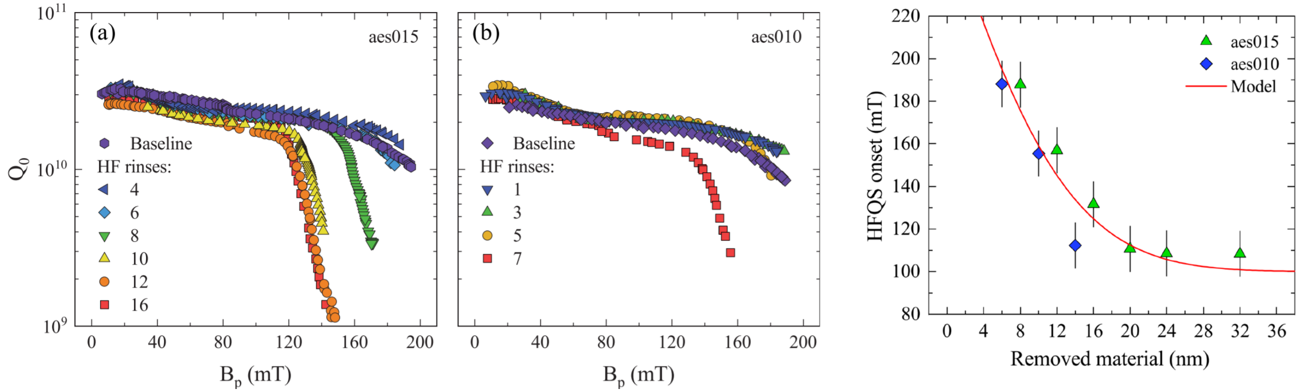


[2] Meissner screening current redistribution





Reappearance of HFQS with HF rinses



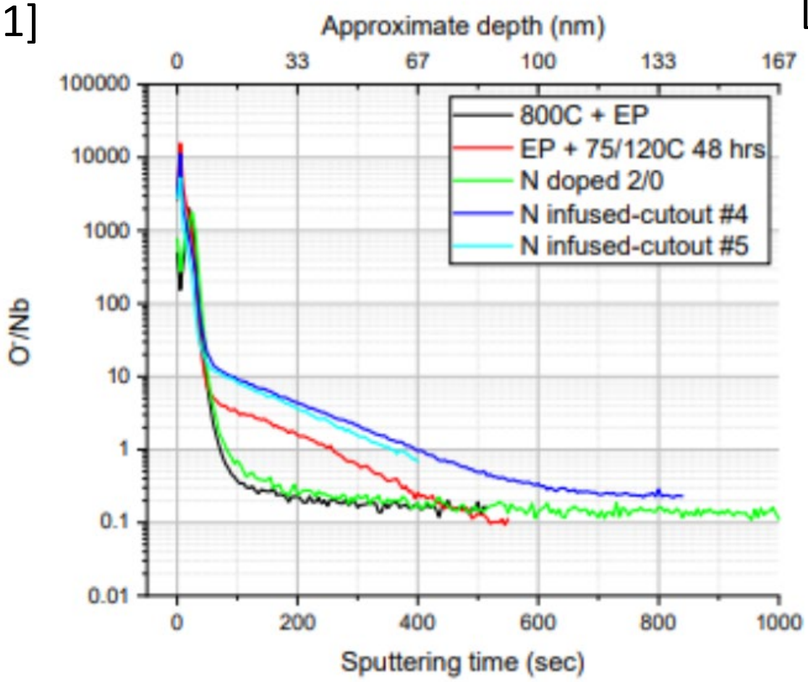
- Many theoretical and experimental results suggest that shallow impurity profiles can be used to extended E_{max}
  - We are currently pursuing this using the oxide dissolution and oxygen diffusion model we have characterized
- Further characterization of Nb samples heat treated with only a temperature ramp to improve O diffusion modeling for short diffusion profiles

[1] T. Kubo *Superconductor Science and Technology* 34.4 (2021): 045006.
[2] M. Checchin and A. Grasselino Appl. Phys. Lett. 117, 032601 (2020)
[3] N, Vudtiwat, and J. A. Sauls. Physical Review Research 1.1 (2019): 012015.

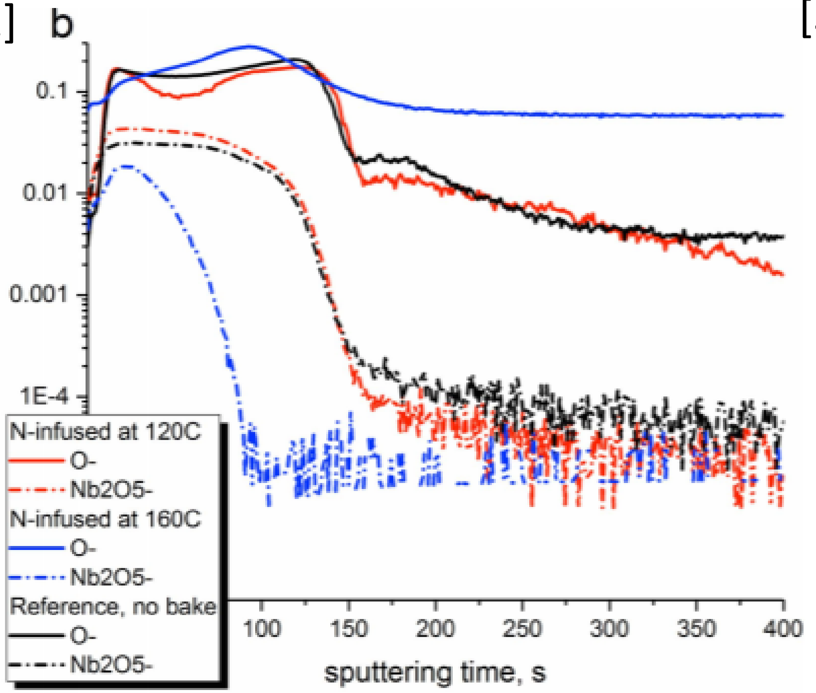
Role of Oxygen in Infusion Recipes

- SIMS measurements show that oxygen may be the dominant agent in “N-infused” cavities

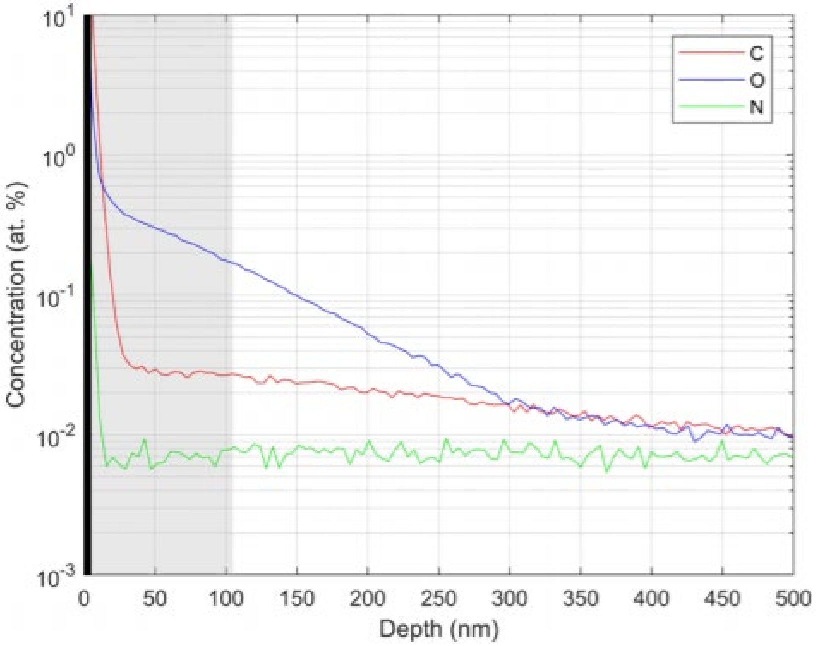
[1]



[2]



[3]

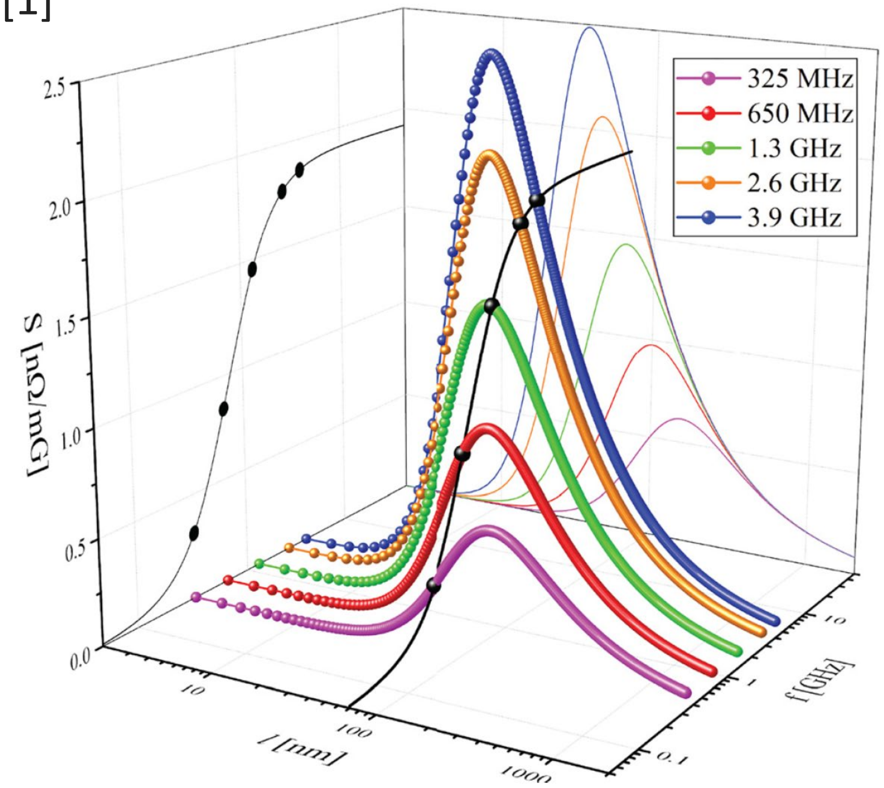


- If the infusion recipe is related to the impurity profile, can the impurity profile in “N-infused” cavities be engineered using only O and allow for a reproducible high Q_0 high, high E_{acc} cavity?

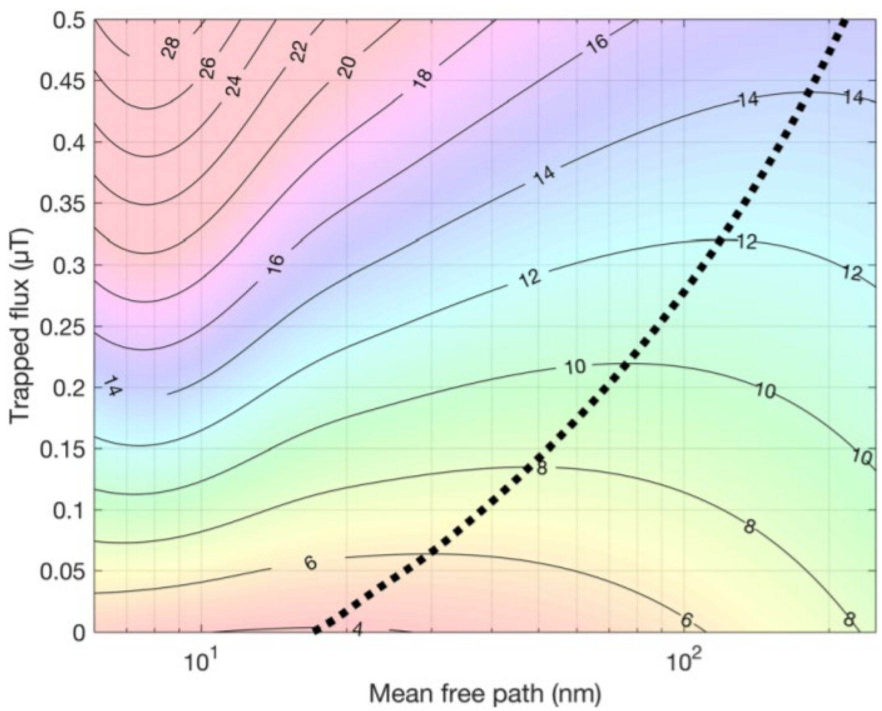
[1] A. Romanenko in *19th International Conference on RF Superconductivity* (JACoW, Dresden, 2019).
[2] A. Grassellino *et al.*, Superconductor Science and Technology **30**, 094004 (2017).
[3] J. Maniscalco, et al., in *19th International Conference on RF Superconductivity* (JACoW, Dresden

Necessity for Impurity Tunability

[1]



[2]



- Sensitivity to trapped flux is frequency and mean free path dependent
- Depending on expected trapped flux, mean free path tunability is required!
- A deeper understanding of the native oxide dissolution and oxygen diffusion process can help guide precise alloying

[1] Checchin, Mattia, et al. *Applied Physics Letters* 112.7 (2018): 072601.
[2] Maniscalco, J. T., D. Gonnella, and M. Liepe. *Journal of Applied Physics* 121.4 (2017): 043910.

Conclusions/Where Next?

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- We've used SIMS to determine parameters in modeling the Nb pentoxide dissolution and O diffusion process.
- Extended Ciovati's model of native oxide dissolution for Nb which may be of use to other oxide dissolution processes.

Where next?

- Continue to explore the oxide dissolution parameter space with cavity test and SIMS analysis
- Explore the role of tailored impurity diffusion profiles within the RF penetration depth
- Explore multiple dissolutions or multi-step temperature profile
- Apply recipes to cavities with a more complex geometries

Acknowledgements

Virginia Tech (SIMS)

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JLab

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Charlie Reece

NCSU

Fred Stevie

JLab Staff

Question Time…

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