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Study of Anode Heating to Reduce Contaminants in the SMP Diode

S. Simpson, R. Goeke, N. Bennett, T. Renk, and D. Nielsen

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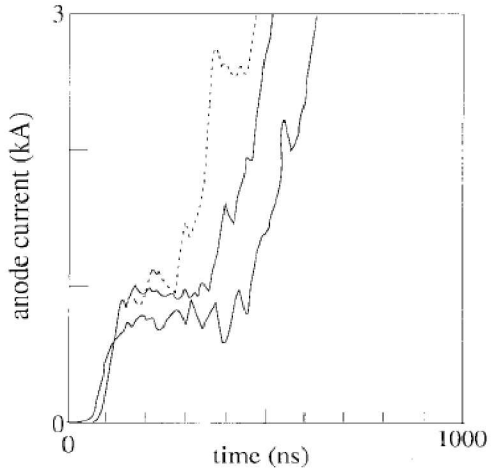
Why In-Situ Heating of the SMP Anode Converter?

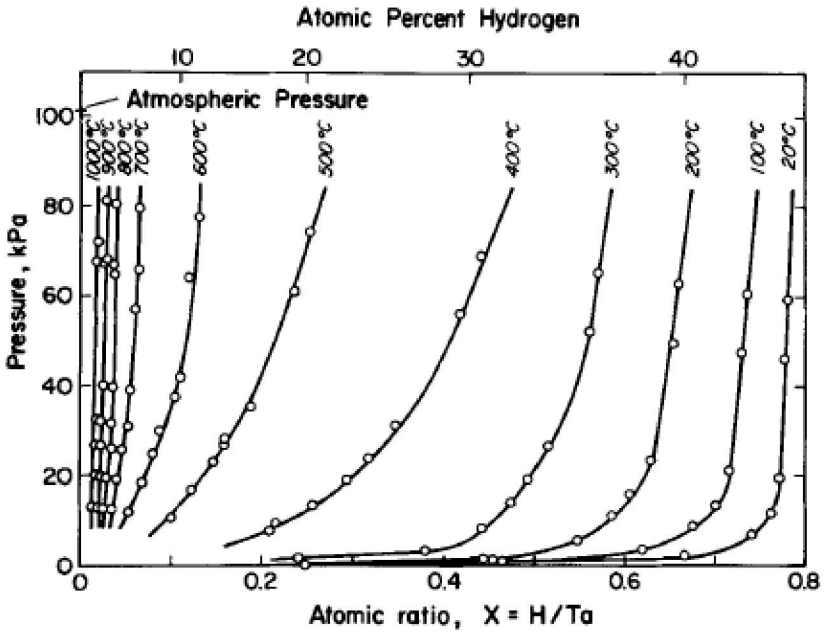
- The Tantalum surface and bulk material consists of contaminants such as H, H_2O, CO, CO_2, N_2, and O_2
  - Monolayers form even in vacuum – requiring in-situ heating to remove contaminants with binding energies > 50 kJ/mol [3,4]
- AK Gap closure rates appear dependent on low-Z back-streaming ions
  - Thought to be dominated primarily by H and H_2O[7]
- Temperatures as low as 650 ºC can be used to remove most of the H and H_2O at pressures of ~10^{-6} torr
  - May improve pulse duration by drastically reducing the number of contaminants which contribute to AK gap closure causing impedance collapse.[8]
- Understanding and controlling the overall surface and bulk contamination should:
  - Reduce risk of premature impedance collapse
  - Improve reproducibility
  - Improve beam spot size
  - Extend the radiographic pulse
  - Increase the overall x-ray dose[4]



Delayed impedance collapse resulting from anode cleaning. Graphic from [8]

What Temps are Required to Clean Tantalum?

- **Isotherms for Ta-H composition indicate that an increase in T while in vacuum results in less H in/on the bulk Ta.**
- **H readily desorbs for T > 650 ºC**
- **CO desorbs for T > 1600 ºC**
- **O_2 and N_2 desorbs for T > 2000 ºC**



Ta-H Pressure Composition Isotherms

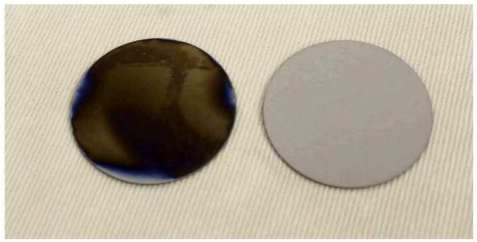
The H-Ta (Hydrogen-Tantalum) System,
A. San-Martin and F.D. Manchester
Journal of Phase Equilibria Vol. 12 No. 3 991

Because tantalum reacts with hydrogen at elevated temperatures, components for electron tubes made of this metal must be outgassed in high vacuum (pressures of 10^{-6} torr or lower). Vacuum outgassing is most rapidly accomplished at 2000°C, although temperatures as low as 1800°C can be used for longer times. The major contaminants of tantalum are oxygen, carbon, nitrogen, and hydrogen. If both oxygen and carbon are present (which is likely to be the case if the metal has been handled without due regard for cleanliness), a reaction between them takes place in the neighborhood of [redacted]. Raising the temperature to 1800°C removes whichever constituent is present in the smaller amount. Very small traces of [redacted]. All outgassing procedures must be preceded by thorough chemical cleaning or degreasing in fresh solvents.

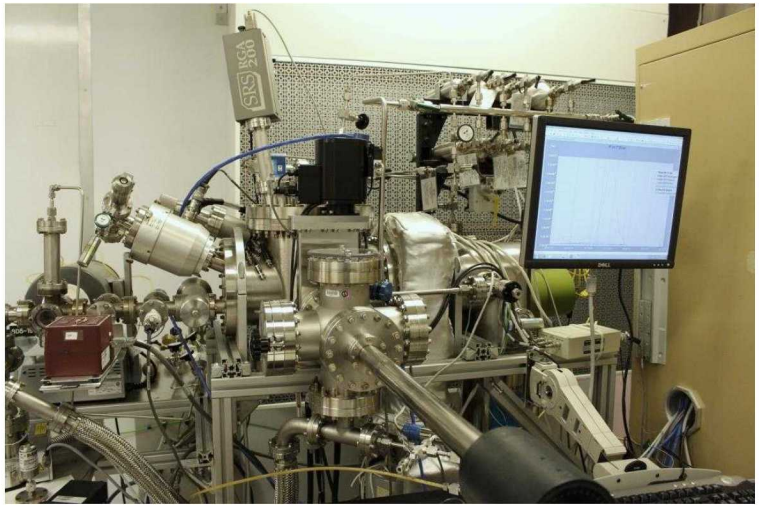
Handbook of Electron Tube and Vacuum Techniques
by Fred Rosebury (Addison-Wesley Publishing, 1977)

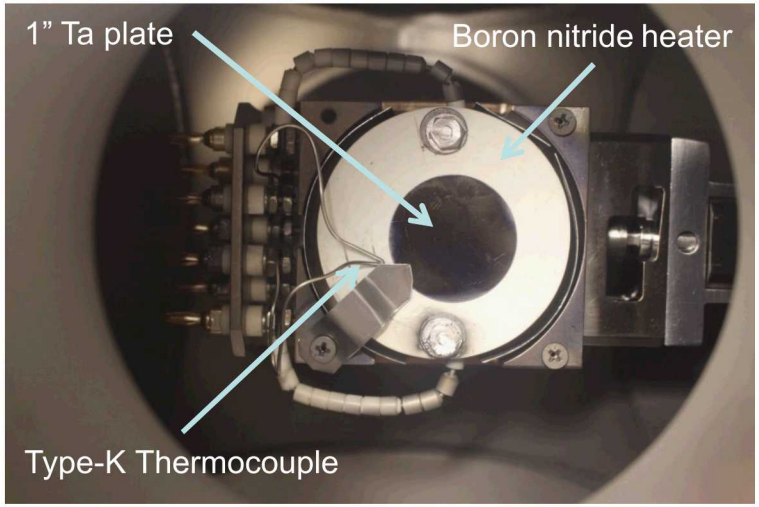
TPD Experimental Setup for Ta Samples

- **UHV Temperature Programmed Desorption (TPD) System**
  - **Capable of 10^{-10} torr**
- **1” diameter Ta samples of 0.015” (.38 mm), 0.040” (1 mm), and 0.060” (1.5 mm) thickness were characterized up to 1100 ºC**
  - **Ta was first cleaned with nitric acid to remove brass residue left behind from the wire EDM process**
- **System is equipped with various calibrated leak sources such as H_2 for calibrating the RGA**
  - **Can calculate the number of moles of H_2 present in/on the bulk Ta prior to heating**



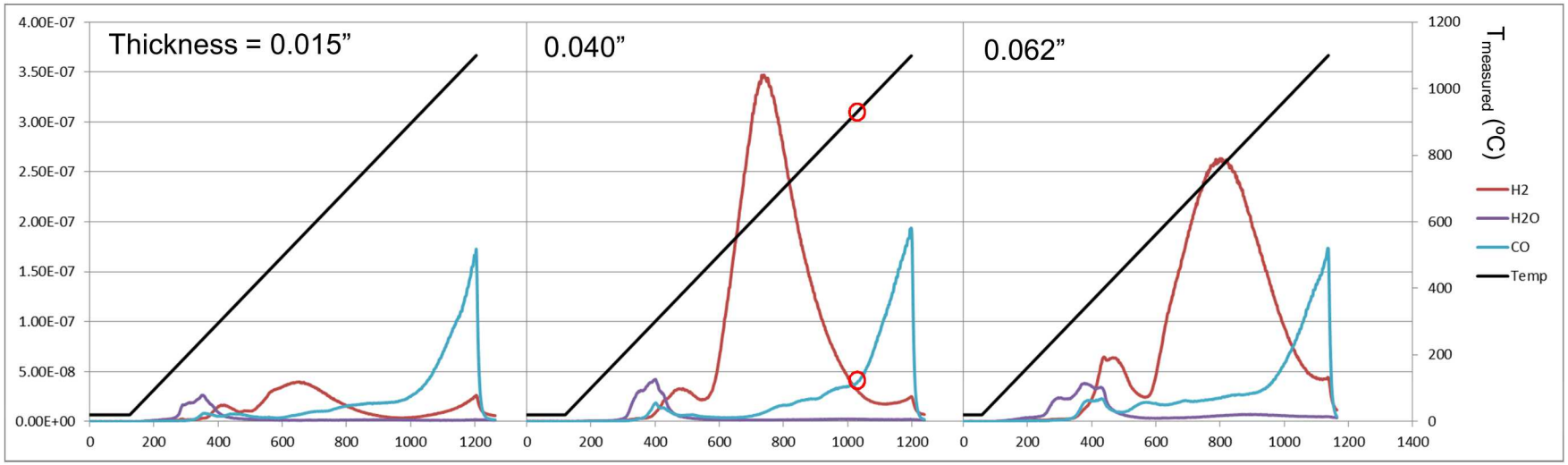
Tantalum sample before and after 1100 ºC bake-out.





TPD Results for Varying Thicknesses of 1” dia. Ta

Partial Pressures for H_2, H_2O, and CO vs. Time



H_2 peak at T= 548 °C
H/Ta = 0.067
~.00119 mols of H

H_2 peak at T= 640 °C
H/Ta = 0.147
~.00696 mols of H

H_2 peak at T= 764 °C
H/Ta = 0.093
~.00683 mols of H

CO beginning to outgas at end of TPD ramp (1100 °C)

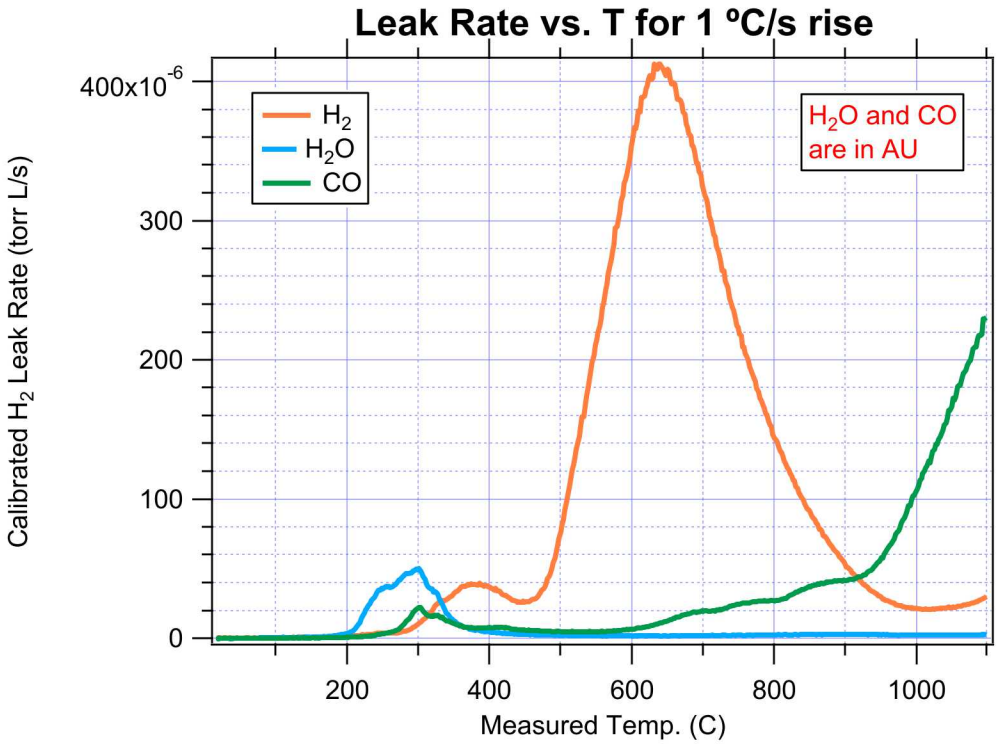
- **First H_2 peak may be surface reactions[10] corresponding to monolayers, or desorption of β-phase Ta-H**
  - **Both of which have lower binding energies**
- **Even with a 1 ºC/s ramp rate, all the H_2O and H are depleted within 1000 s**
- **Nearly equivalent H_2O and CO curves indicates these are evolving from the surface**
- **CO starts to desorb fairly rapidly for T > 925 ºC**
  - **Desorption of CO may be responsible for the increase of H at t = 1000 s as the CO reacts with the H_2O on the chamber walls releasing CO_2 and H_2**
  - **Doing a TPD with a much slower temperature ramp rate will reveal whether the late H_2 is coming from the sample or a chemical exchange with the chamber walls**

TPD Results: Calculating Binding Energies for Ta-H

- **TPD Analysis using Redhead [5,6]**
  - **Allows for extracting activation energies from a single desorption spectrum**
  - **Assumes that the activation parameters are independent of surface coverage**
  - **Assumes desorption follows 1st order kinetics (H_2 desorption is actually a 2nd order process), but for an estimate we can use:**
    - $\Delta E_{des} = RT_{max}\left[ln\frac{v_1 T_{max}}{\beta} - ln\frac{\Delta E_{des}}{RT_{max}}\right]$ **Eq. 1**
    - ΔE_{des} is binding energy (J/mol), R is gas constant (8.3 J/[mol K], T_{max} is temperature of max desorption (K) i.e. $\left.\frac{dr_{des}}{dT}\right]_{T_{max}} = 0$, v_1 is rate constant for desorption for first order (s^{-1}), β heating rate (K/s)
    - $ln\frac{\Delta E_{des}}{RT_{max}}$ is estimated as 3.64; v_1 is typically chosen as 10^{13}/s
- Yields binding energies for H on our Ta samples somewhere between 231 - 294 kJ/mol depending on sample thickness
  - Others have measured 389 kJ/mol for H on Ta[9]
  - Discrepancy most likely results from:
    - Using Eq. 1 which is only an approximation
    - Other contaminants present can reduce the effective binding energy

TPD Results: Desorption Equivalent Leak Rates vs. Temperature for 0.040” Ta

- Integrating the corrected H_2 leak rates yielded 64 torr L
  - Using the Ideal Gas Law, the number of moles H_2 is given by:
    - $n = \frac{PV}{RT} = .00348\ mols$ of H_2
    - Or equivalently .00696 mols of H
  - At standard temperature and pressure, this yields ~82 mL of desorbed H_2 from a 1” dia. Sample
  - During rapid heating, this could result in a plasma density of ~1x10^{17} cm^{-3} in a 10 mm AK gap[3]
- H_2O is completely desorbed from the surface by 400 ºC
- H_2 is desorbed by 1000 ºC
- CO may completely desorb if given enough time



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