Tritium on Metal Surfaces

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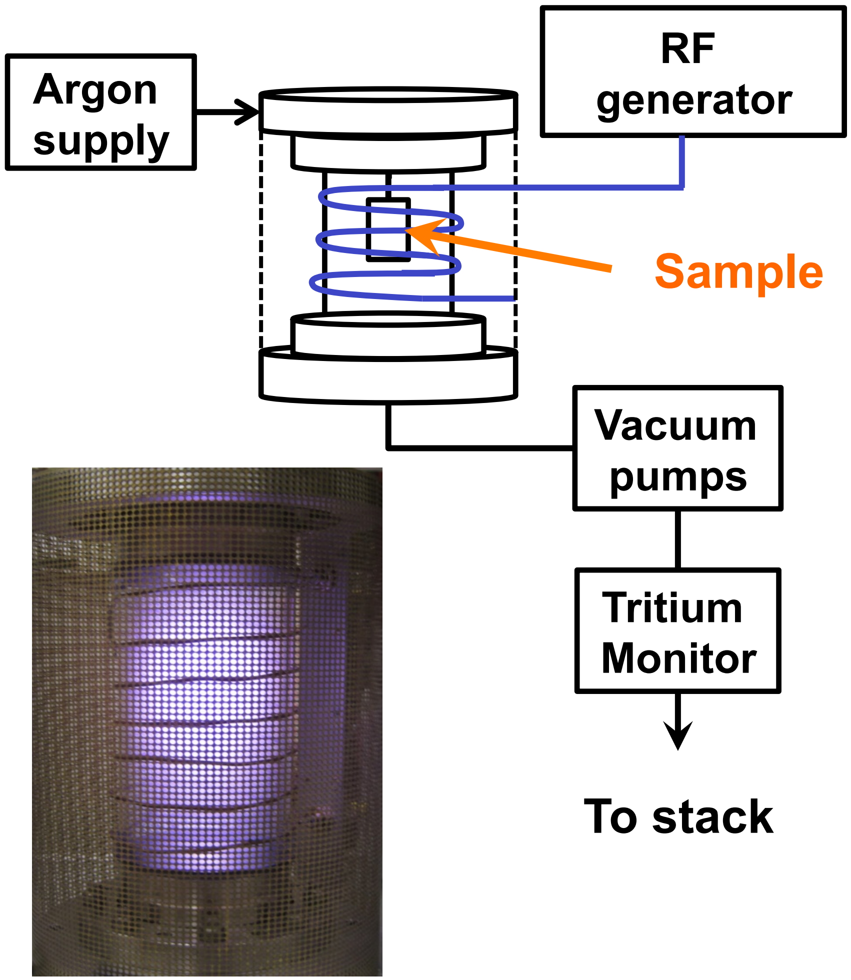
Idaho Falls, Idaho
Sept 23-25, 2014

Summary/Conclusions

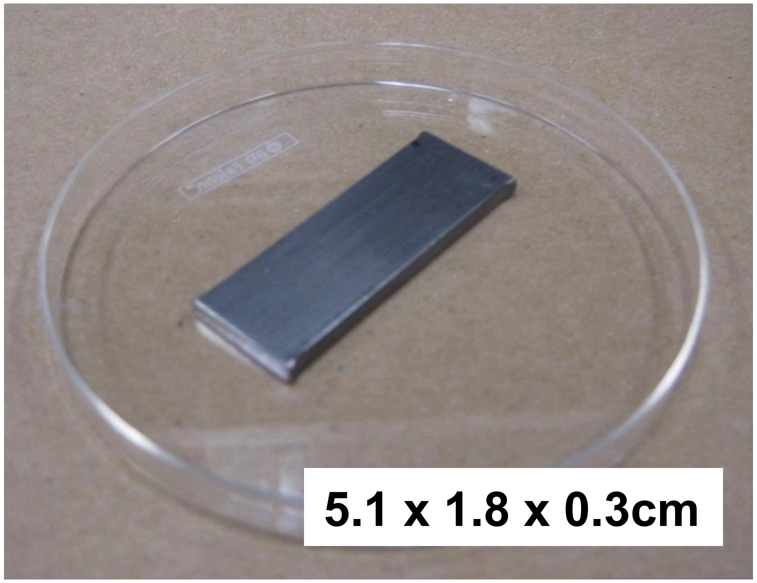
- Tritium concentrates in the water layers covering metal surfaces
- Water layers on the surface 'pump' tritium from the metal bulk
- The relative removal rate of tritium from the surface does not depend on
  - the initial tritium loading pressure at room temperature,
  - the storage time in an inert environment, or
  - the metal type
- Regrowth of surface activity is:
  - rapid, and
  - controlled by diffusion from the 'near-surface' bulk

An argon plasma generated by a radio-frequency (RF) field was used to desorb tritium from metals into a flowing gas stream



- Plasma was ignited in argon by passing a 13.56 MHz AC current through a copper coil
- Sample floated at the plasma potential
  - → ionic flux = electron flux
- Tritium released from the sample was purged into a downstream in-line tritium monitor
- Base pressure ≈ 10^{-4} Torr
  - → Trace water in vacuum system re-deposits on metal surface within 15 sec

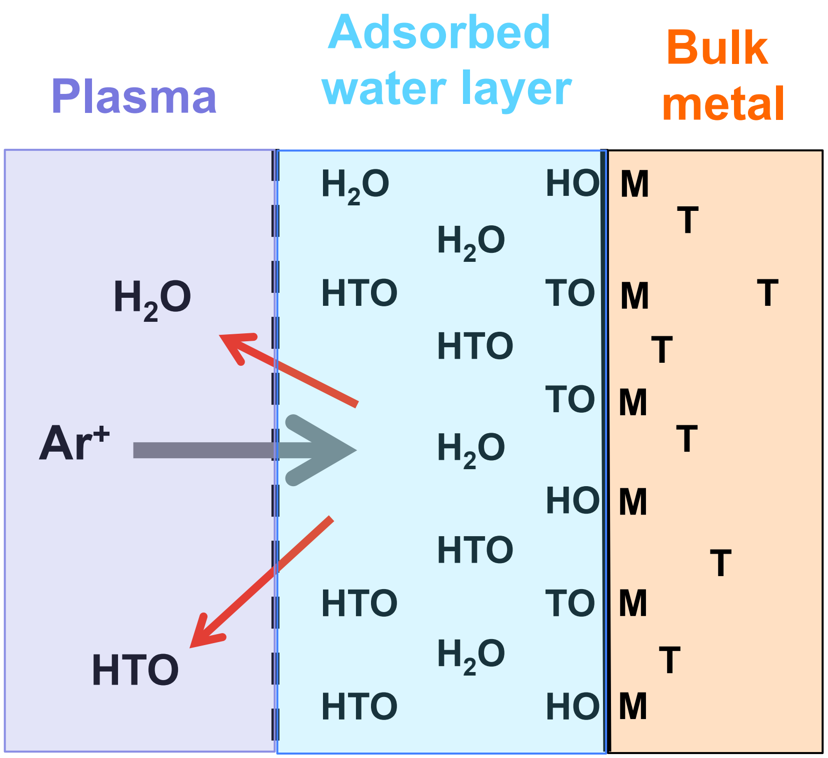
Metallic samples were de-greased, stored in hard vacuum for 24 hours, then charged with DT gas at room temperature



- Samples were separated from each other during the DT loading
- Samples stored under -50°C DP helium until experiment
- Batch #1 stored in same container & removed using a glove-bag
- Batch #2 stored in separate containers

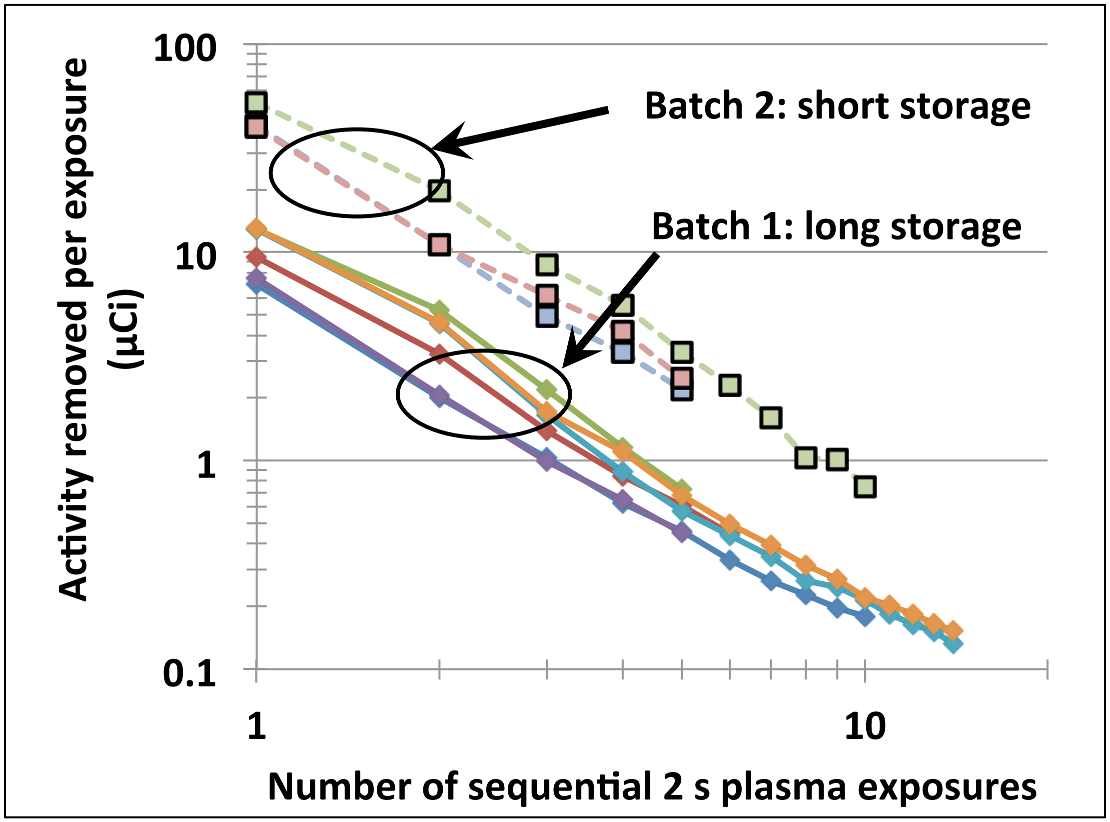
Batch Number	Time (hrs)	Pressure (Torr)	Isotopic Ratio (%)	Storage time
1	3	687	45%	3.5 years
2	24	659	39%	36 days

Water was removed from the sample surface using a series of 2-second plasma bursts



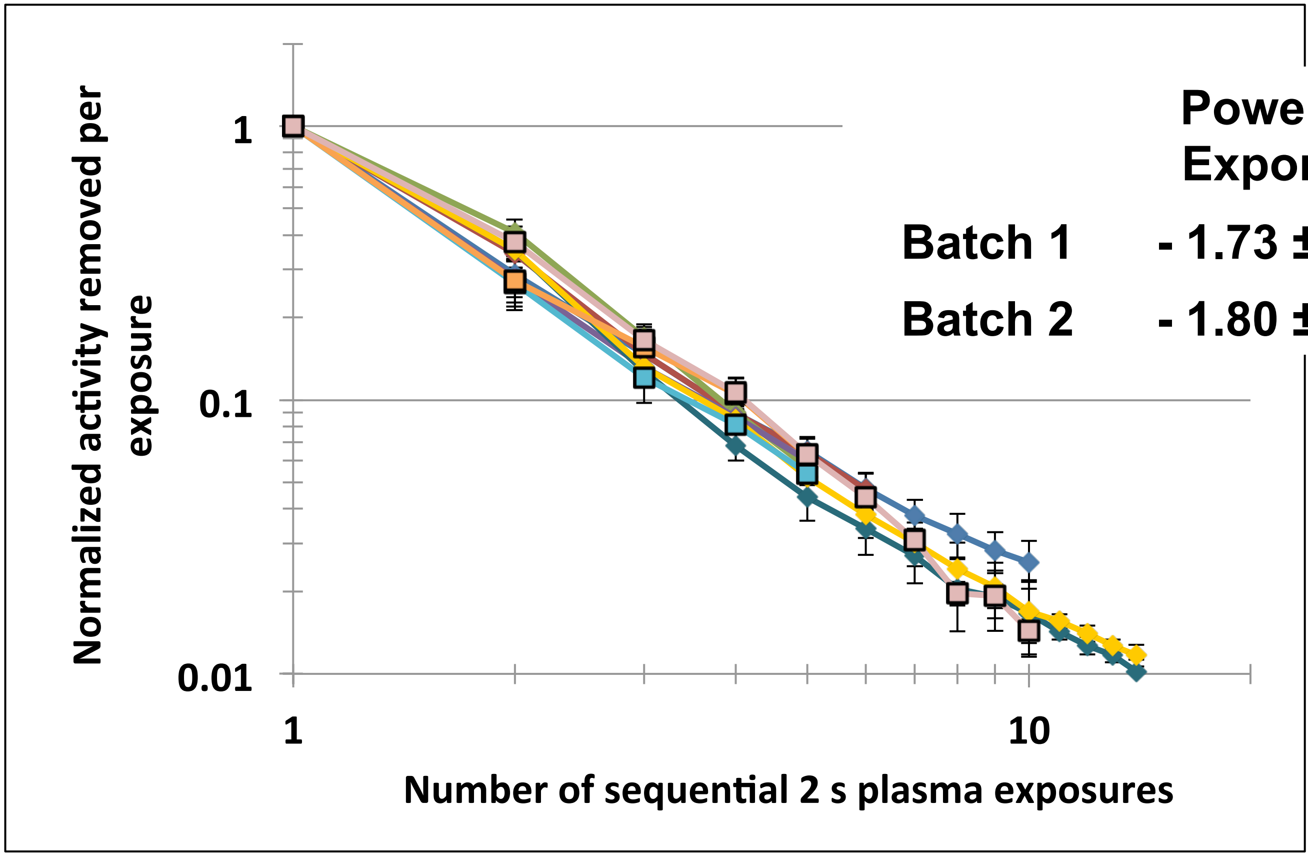
- Metal surface comprises:
  - Hydroxyl layer
  - Proton bonded ‘ice’ layer
  - Mobile Van der Waal bonded water
- Adsorbed water layer regrows between exposures
- Tritium migrates into freshly formed tritium-free water layer

Initial activity on the stainless steel samples determines the amount of activity removed during each following exposure



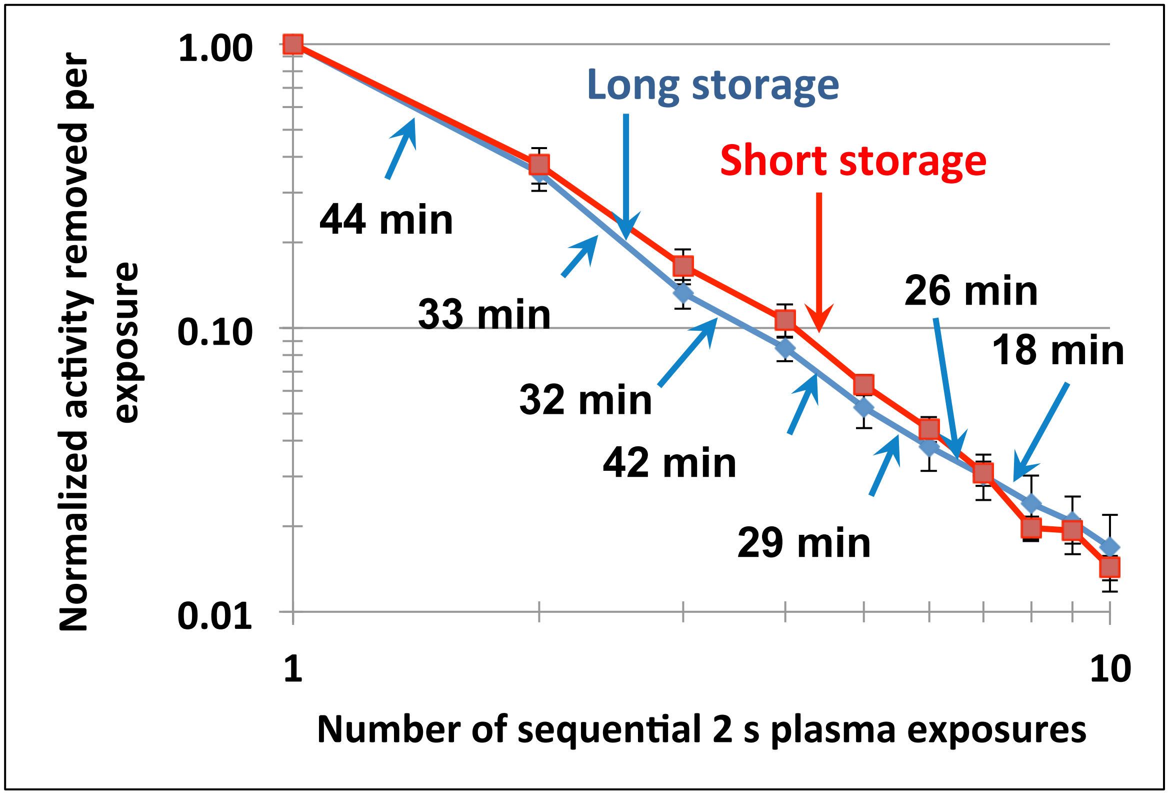
- **Batch 2 samples contain more activity: Shorter storage & longer loading time**

Trend in activity removed does not depend on sample history



- Data were normalized to the initial amount removed
- Trend fitted to a power law

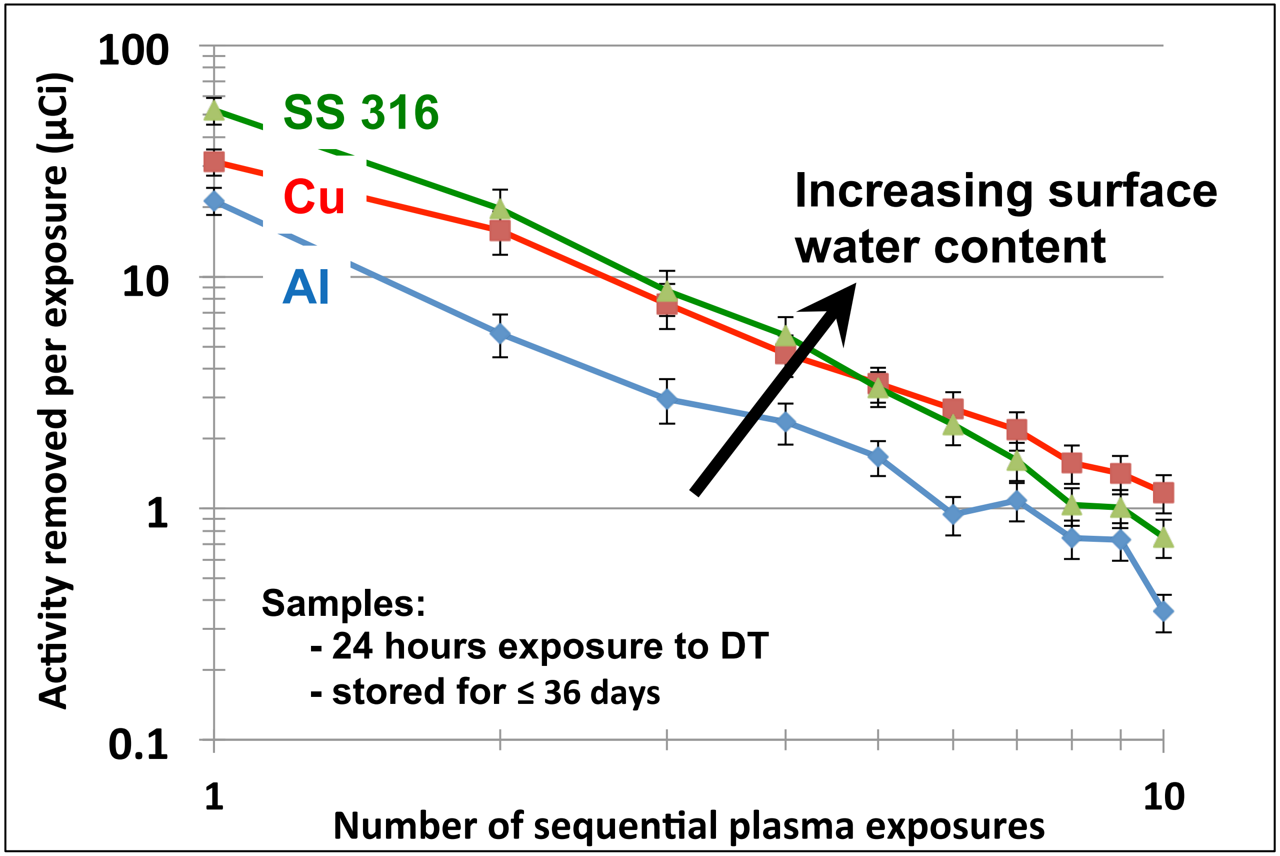
The trend in activity removed does not depend on dwell period between plasma shots



- Dwell times for “Short storage” samples were fixed at 20 min between shots
- Dwell times for “Long storage” samples were varied

Adjusting the dwell times between 18 and 44 min does not change the relative amount of activity removed from the metal

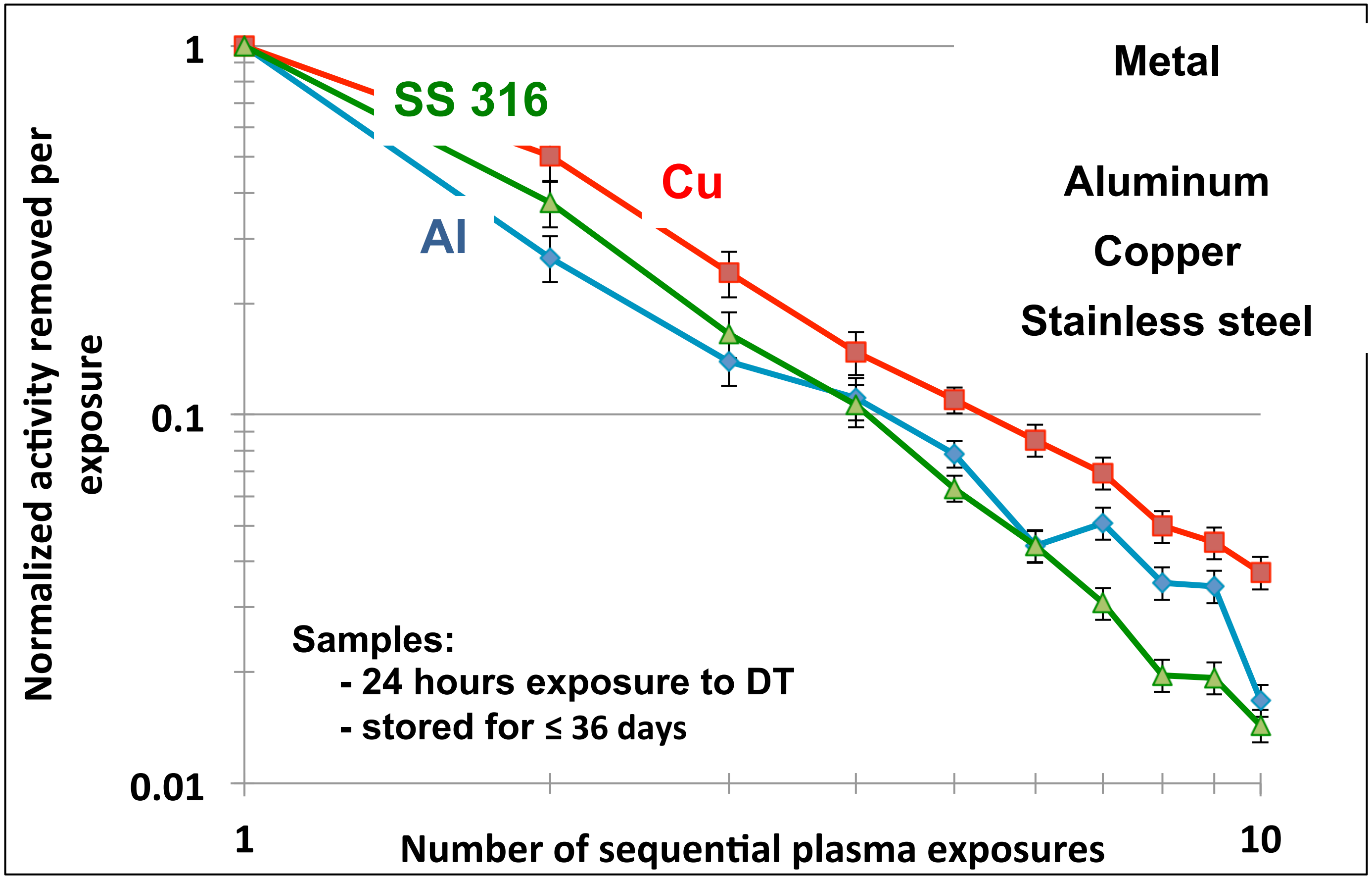
Total removable surface activity increases with the number of monolayers of adsorbed water at a fixed relative humidity



Water isotherm references

- Aluminum: Al-Abadleh, H.A, et al.; *Langmuir*, 19, 2003, p. 341
- Copper: Sharma, S.P.; *J. Vac. Sci. Tech.* 16(5), 1979, p. 1557
- Stainless steel: Ohmi, T. et al.; *Rev. Sci. Instrum.,* 64(9), 1993, p. 2683

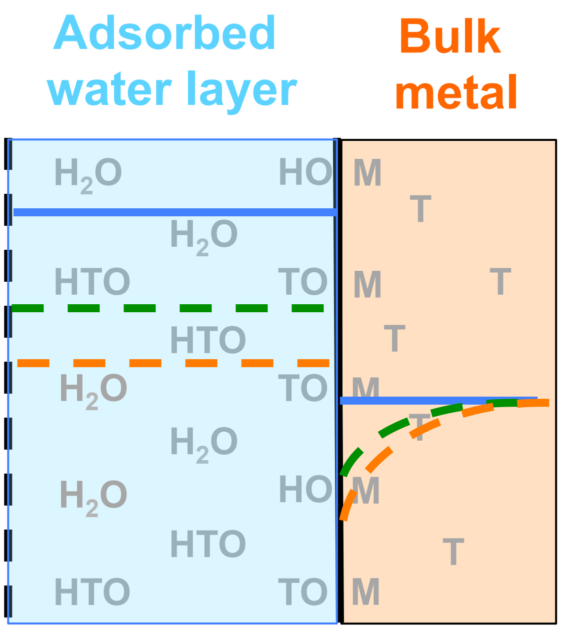
Activity removal from stainless steel, copper, and aluminum appears to follow the same trend for the three metals



Metal	Power law Exponent
Aluminum	- 1.60 ± 0.09
Copper	- 1.47 ± 0.04
Stainless steel	-1.80 ± 0.10

- Data were normalized to the initial amount removed
- Trend fitted to a power law

A model based on Fickian diffusion of atomic hydrogen through two metallurgically bonded media explains the data



$$\frac{c_{water}}{S_{water}} = \frac{c_{metal}}{S_{metal}}$$

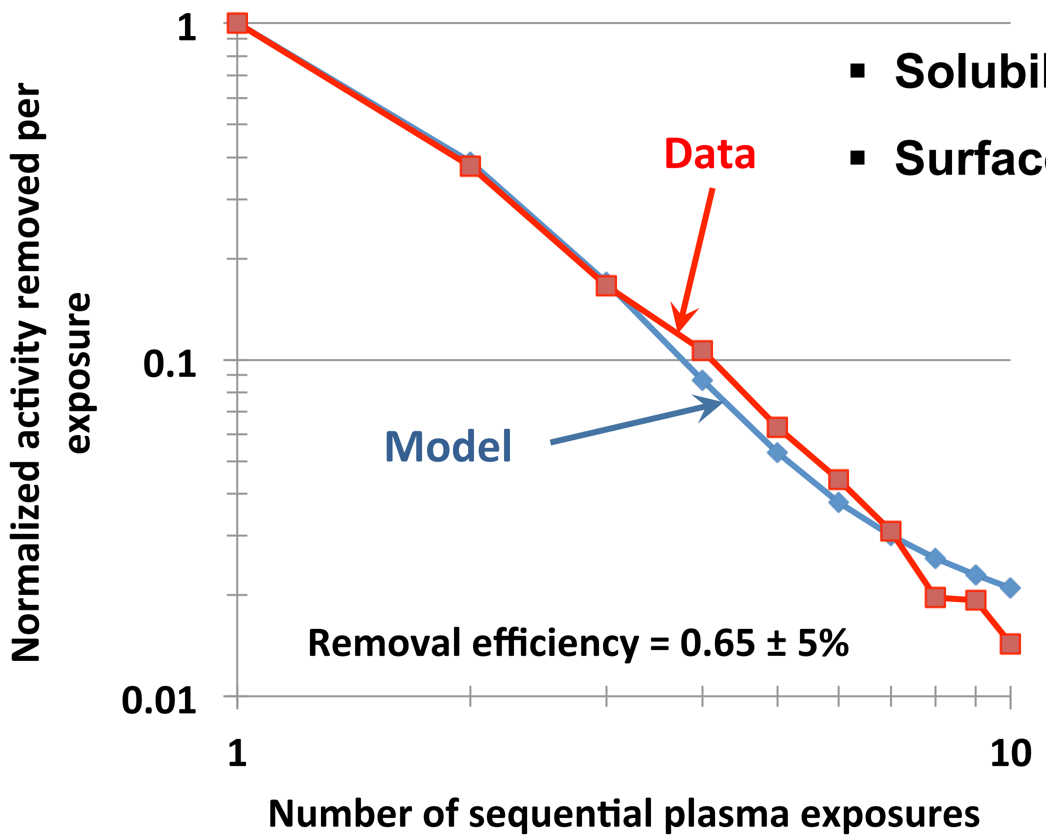
Assumptions:

- **Constant chemical potential across the boundary**
- **Rapid diffusivity through the oxide into the water layer**
- **Tritium migration across metal/oxide boundary limited by metal diffusivity**
- **No tritium loss from sample in storage**

Equilibrium

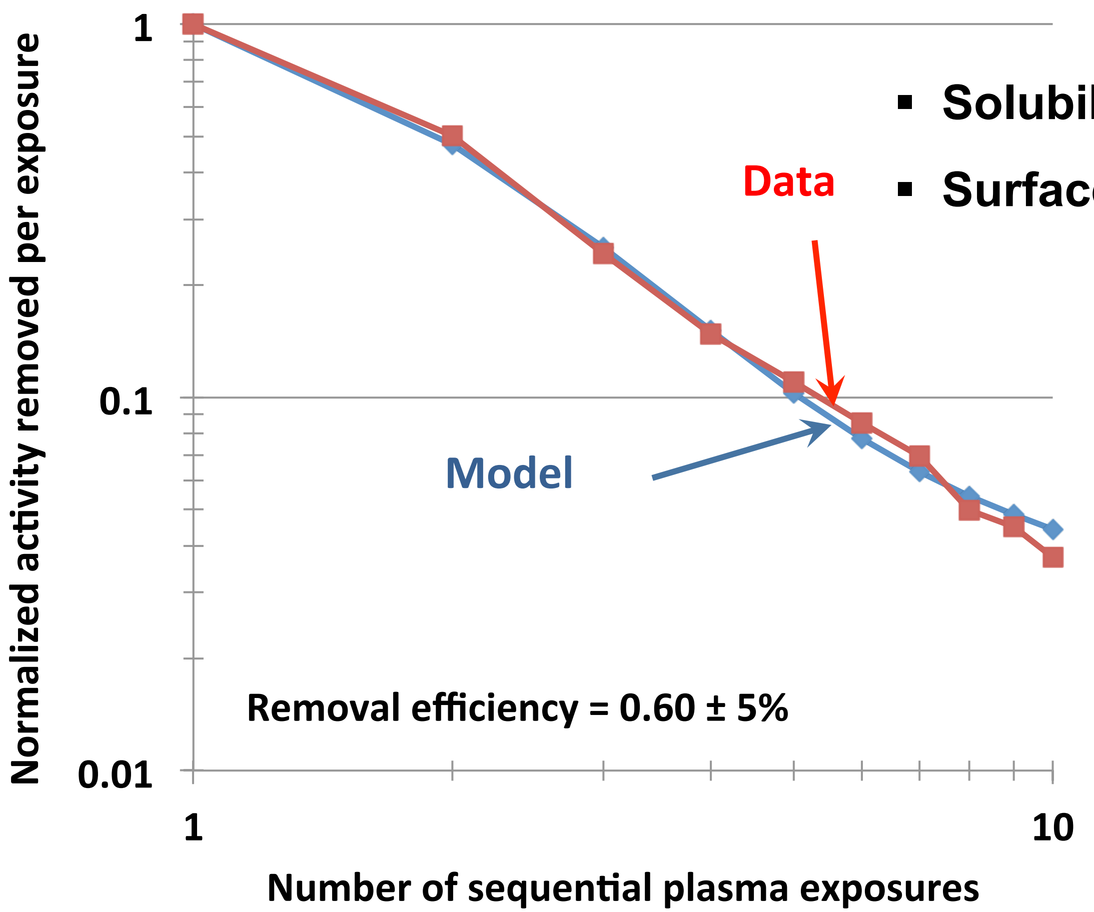
Response to 'T' empty water layer

Less than 1% of surface sites on stainless steel are occupied following an exposure to tritium gas at room temperature



- Solubility ratio = 775 ± 25
- Surface solubility = 1030 mol/m^3

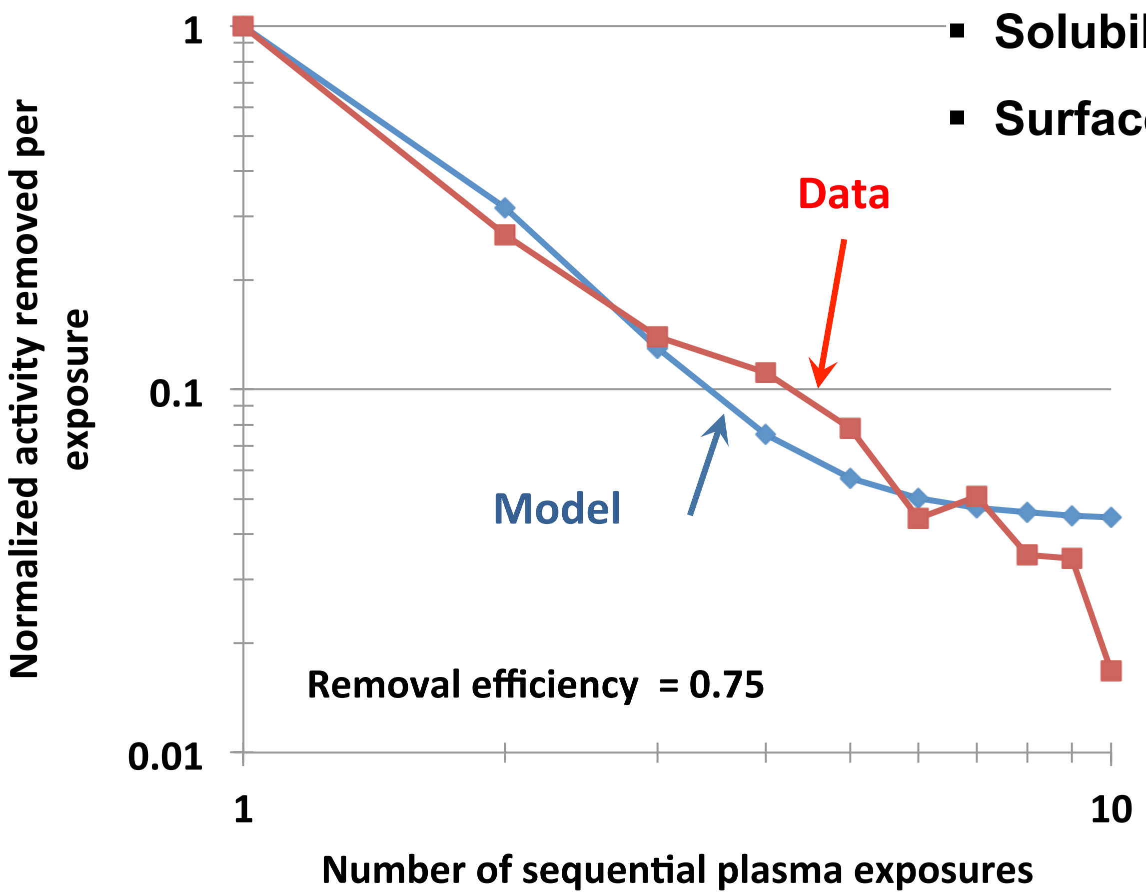
Less than 10^{-3}% of surface sites on copper are occupied following an exposure to tritium gas at room temperature



- **Solubility ratio = 10,500 ± 500**
- **Surface solubility = 1.13 mol/m^3**

Less than 10^{-2}% of surface sites on aluminum are occupied following an exposure to tritium gas at room temperature

- Solubility ratio = 75 ± 25
- Surface solubility = 2.0 mol/m^3



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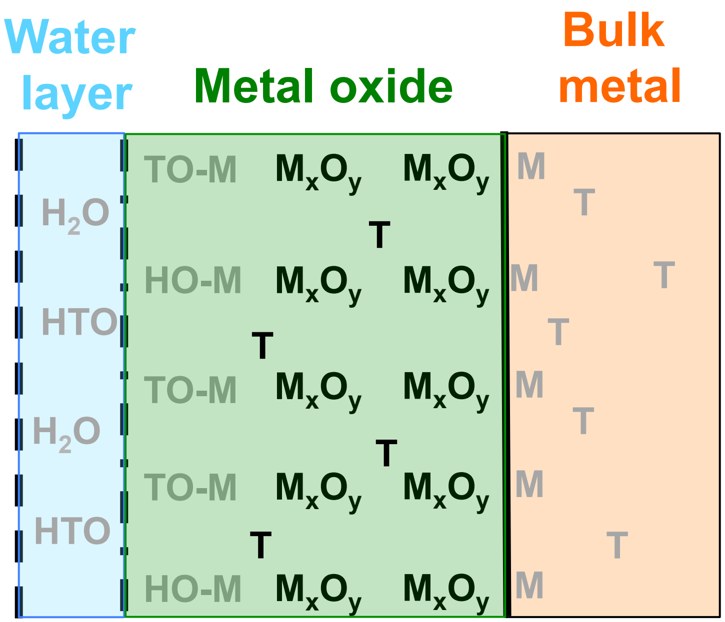
Including the thickness of metal-oxides on metals leads to similar solubilities for tritium on the surface of simple metals: Cu & Al



- Metal-oxide thicknesses grown at room temperature are metal dependent and less soluble than adsorbed water layer
- Surface solubility of Al and Cu including their metal-oxide layers are similar
- Solubility on steel remains high due to complex surface??

RH ~ 0%			Surface Solubility (mol/m^3)	
Metal	Adsorbed water (nm)	Metal-oxide (nm)	Without oxide	With oxide
Al	0.3	1.0	$(4.6 \pm 0.3)\times10^4$	2.0 ± 0.7
Cu	0.3	9.5	36 ± 1	1.1 ± 0.1
SS	0.7	12.0	$(6.2 \pm 0.1)\times10^3$	1030 ± 30