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Investigation of Anomalous Hydrogen Production from Water Adsorbed to Metal Oxide Surfaces

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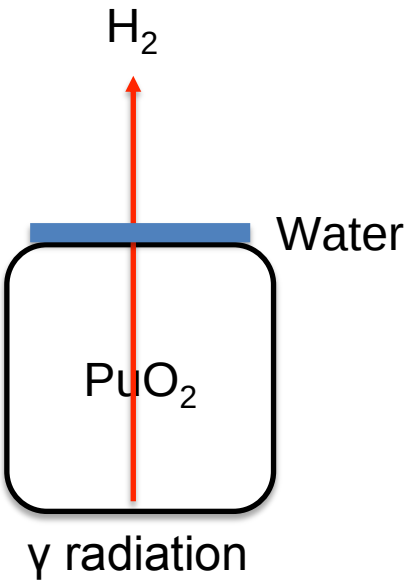
^{60}Co gamma irradiator
- as seen on the BBC!

Why is this Important?

- Plutonium stored in sealed metal containers at Sellafield
- Canisters found pressurised
- H_2 determined to be major component of gas
- Presence of metal oxide has catalytic effect on the yield of H_2
- Caused pressurisation of canisters
- Find metal oxide with similar catalytic activity to PuO_2 to model how stored container will behave over lifetime



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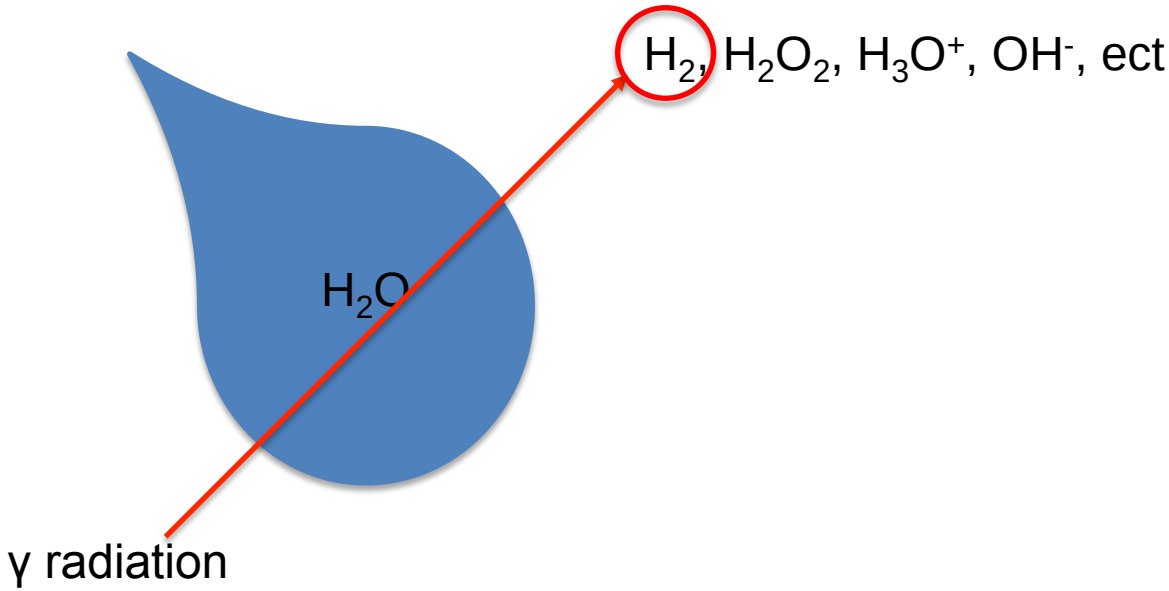
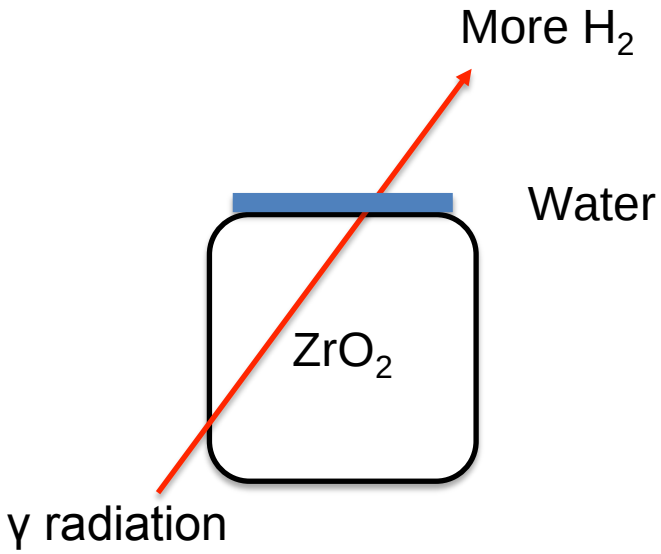
Radiation Chemistry of Water

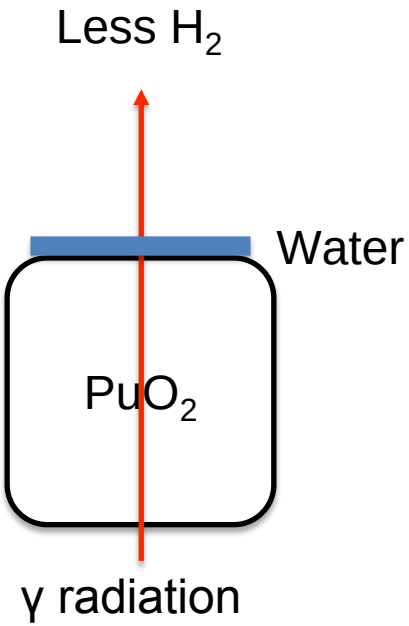
- G value: number of molecules produced per 100 eV of energy input
- Pure water: $G(H_2) = 0.45$ molecules/100 eV



Affect of Metal Oxide

- Metal oxide can increase or decrease $G(H_2)$ compared to pure water
- Or have no effect
- Exact mechanism not known

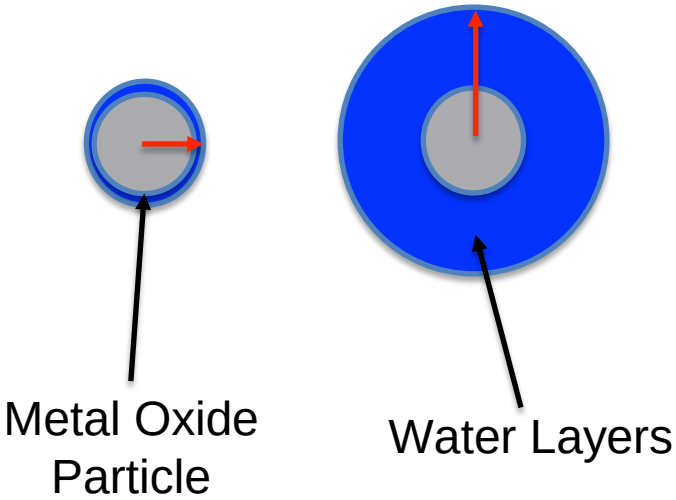




Other Contributions to Increase in $G(H_2)$

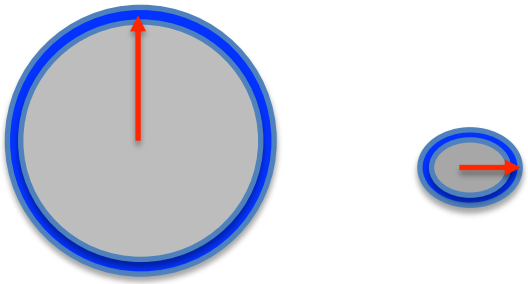
- $G(H_2)$ is greater for lower water loading

Energy must migrate to outer layers for H_2 to be seen



- Greater $G(H_2)$ for smaller particle size

Deposited energy has greater distance to migrate before getting to H_2O



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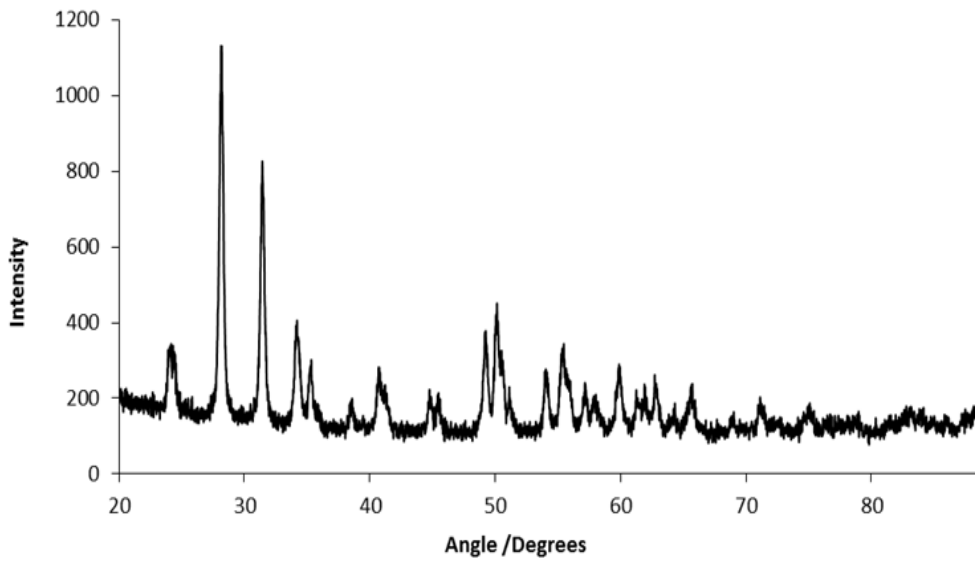
Experimental Method

- H_2 generation from ZrO_2
- Powders baked at 500°C for 24 h when received
- Crystal structure determined by XRD



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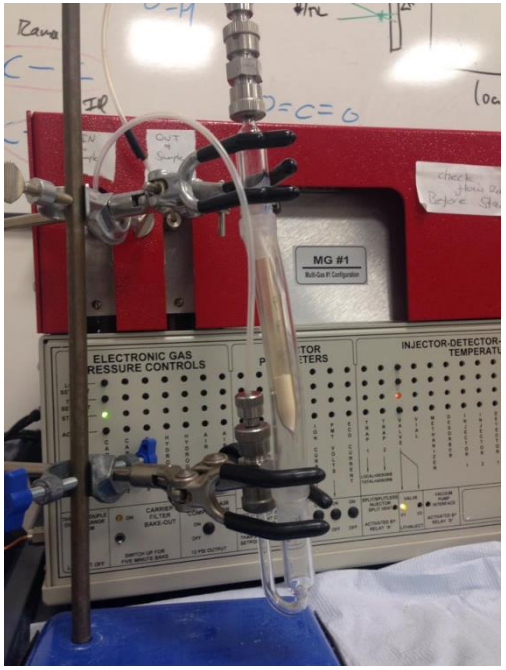
Experimental Method

- Surface area determined by BET
- H_2O introduced to ZrO_2 powder by humidity control
- Salt solution determines %RH of chamber
- Irradiated samples analysed by Gas Chromatography



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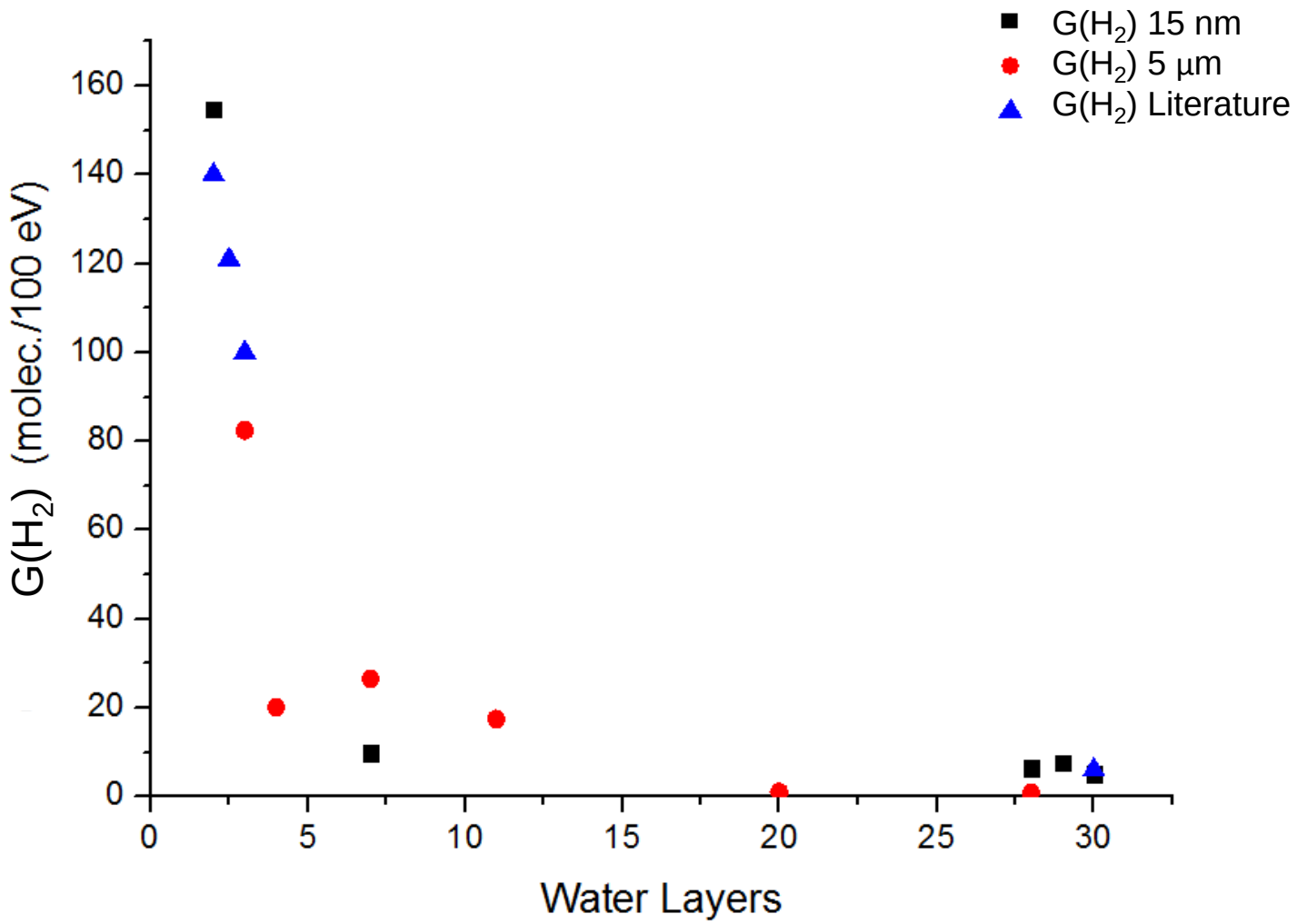
Results



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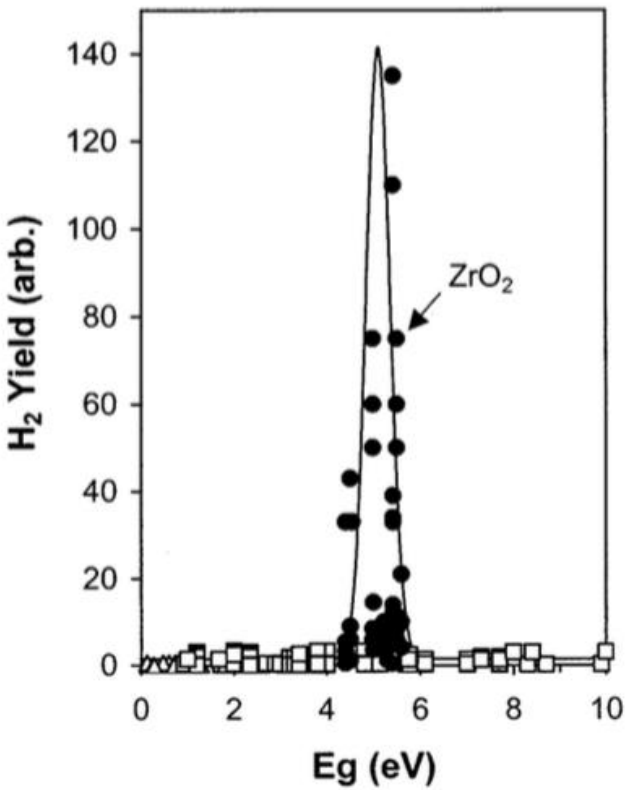
Future Work

- H_2 measured from ZnO as a function of:
  - Water Loading
  - Particle Size
  - Crystal Structure

- Single crystal of ZnO in high vacuum chamber
- Irradiated with 100 eV electrons
- Products detected by QMS
- Alternate ice layers of H_2O and D_2O

Possible Mechanism for $G(H_2)$ Increase?

- Exact mechanism not known
- Band gap has effect upon yield
- Peak in $G(H_2)$ at $E_g \approx 5$ eV
- BDE (H-OH) ≈ 5 eV
- Radiation energy deposited in metal oxide
- Energy transfer from the metal oxide into adsorbed molecule
- Migrates through interface
- Recombines in water molecule – causes dissociation