

Radiolysis of H₂-O₂ Gas Mixtures of Relevance to Long Term Storage of PuO₂

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Outline

- Project Background
- Research Objectives
- Results and Discussion
- Further Work

Background

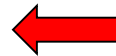
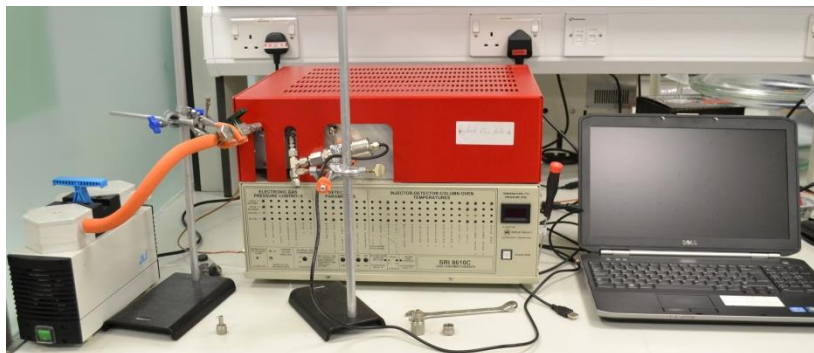
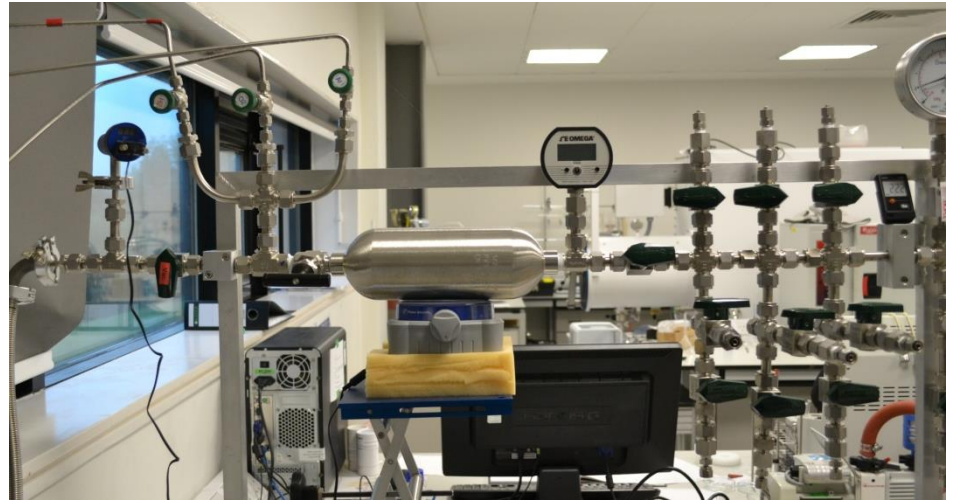
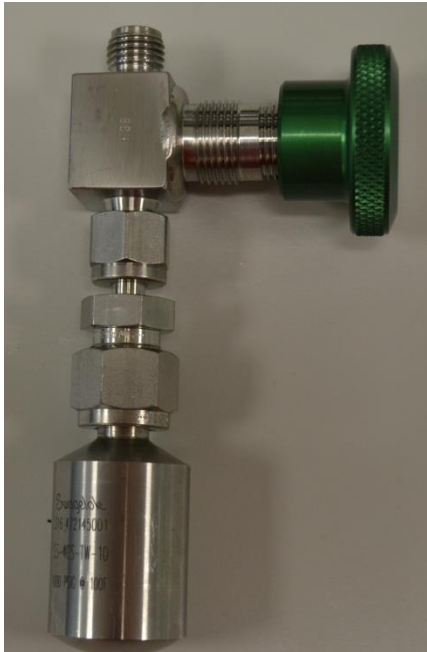
- 120 tonne stockpile of civil separated Pu currently in interim storage at Sellafield
- Stored in multi-can system as PuO_2 powder
- Several mechanisms may lead to canister pressurisation
- Radiolysis of adsorbed moisture may lead to H_2 and O_2 atmospheres



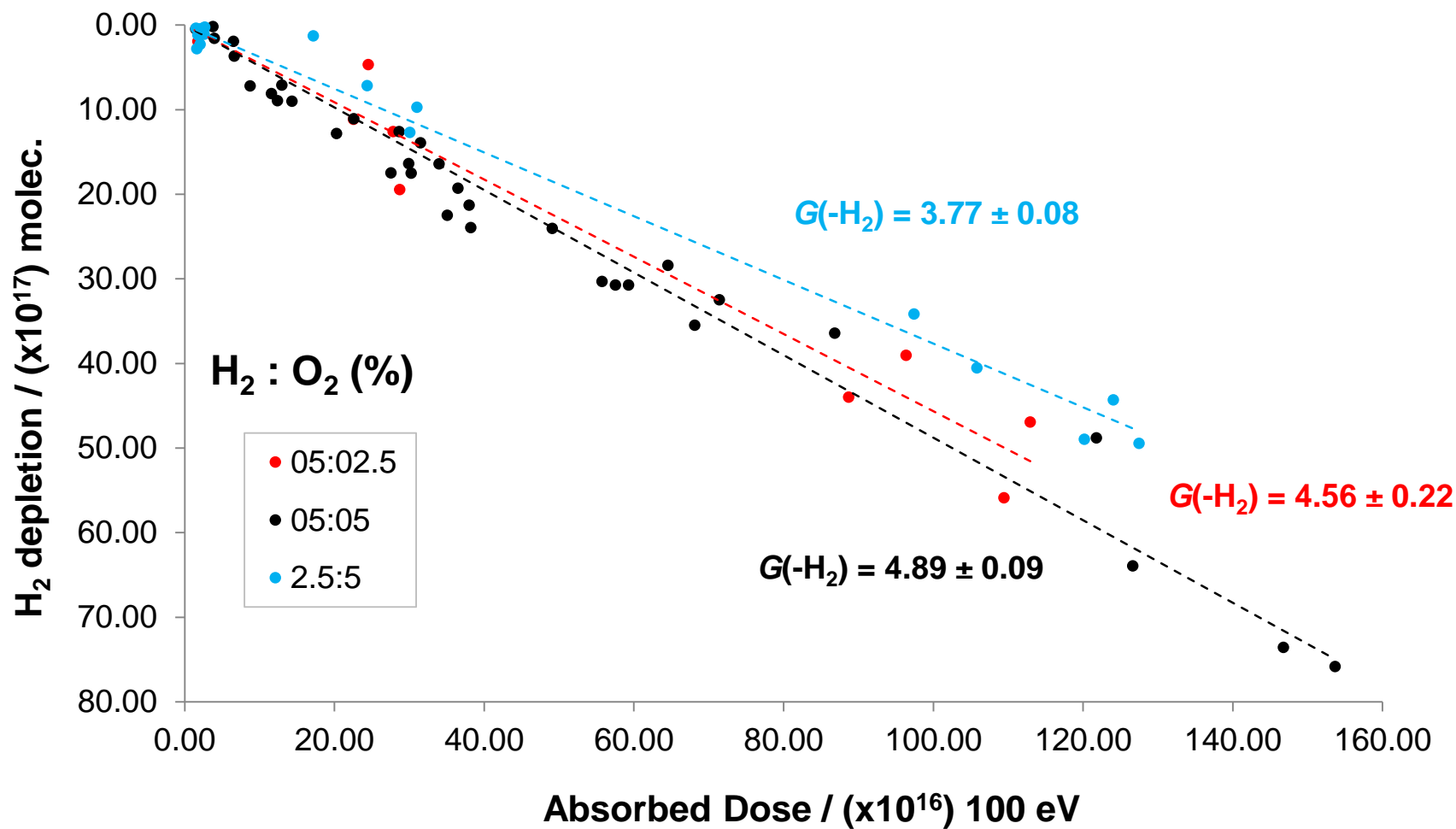
Research Objectives

- Investigate radiation chemistry of $\text{H}_2\text{-O}_2$ gas mixtures
 - 2:1 $\text{H}_2\text{:O}_2$ water stoichiometry
 - 1:1 $\text{H}_2\text{:O}_2$ equal volumes
 - 1:2 $\text{H}_2\text{:O}_2$ excess O_2
- Investigate the effects of PuO_2 surrogate material (CeO_2 and ZrO_2)
- Utilising ^{60}Co γ -rays and 5.5 MeV He^{2+} accelerated ions

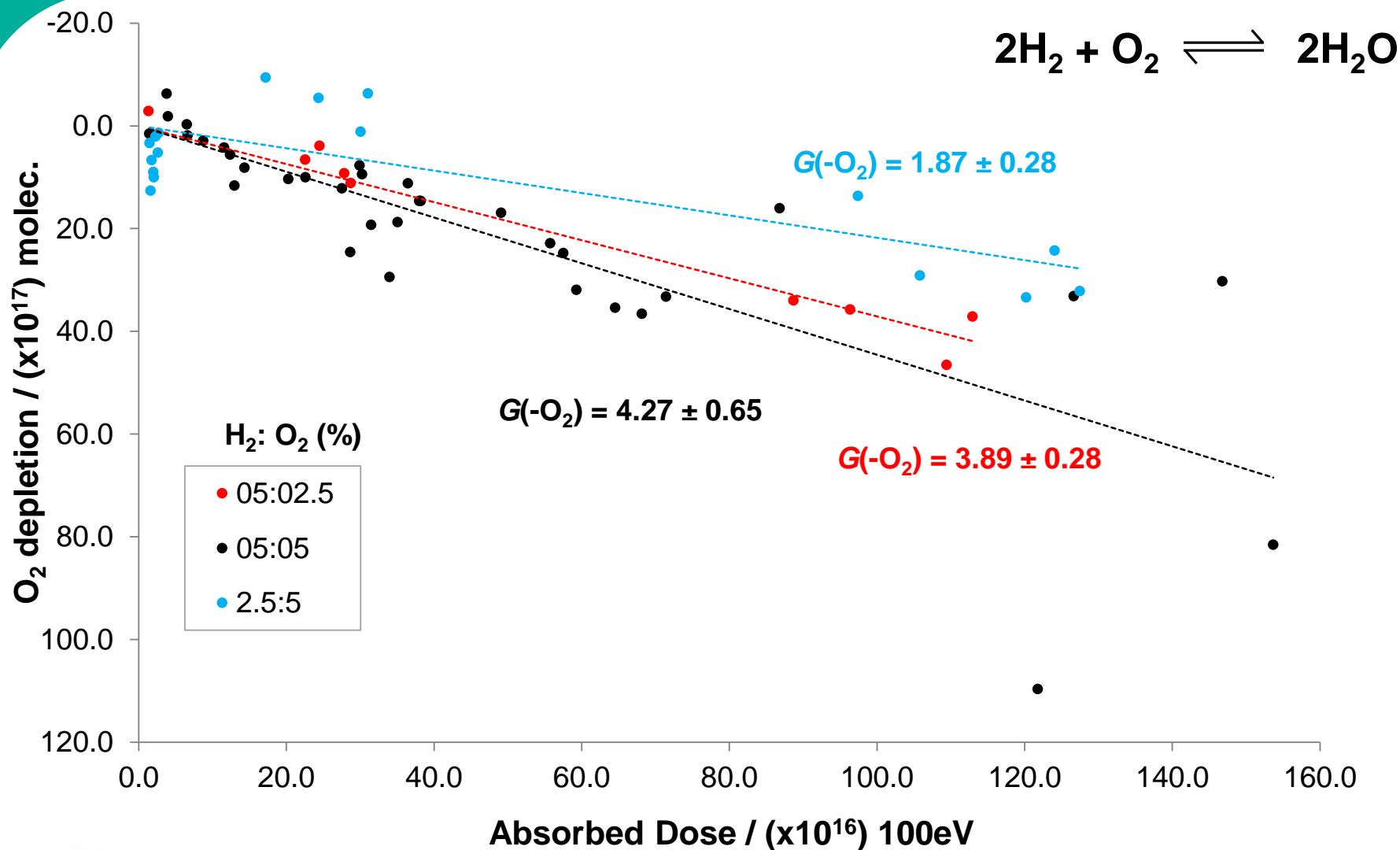
Experimental – γ source



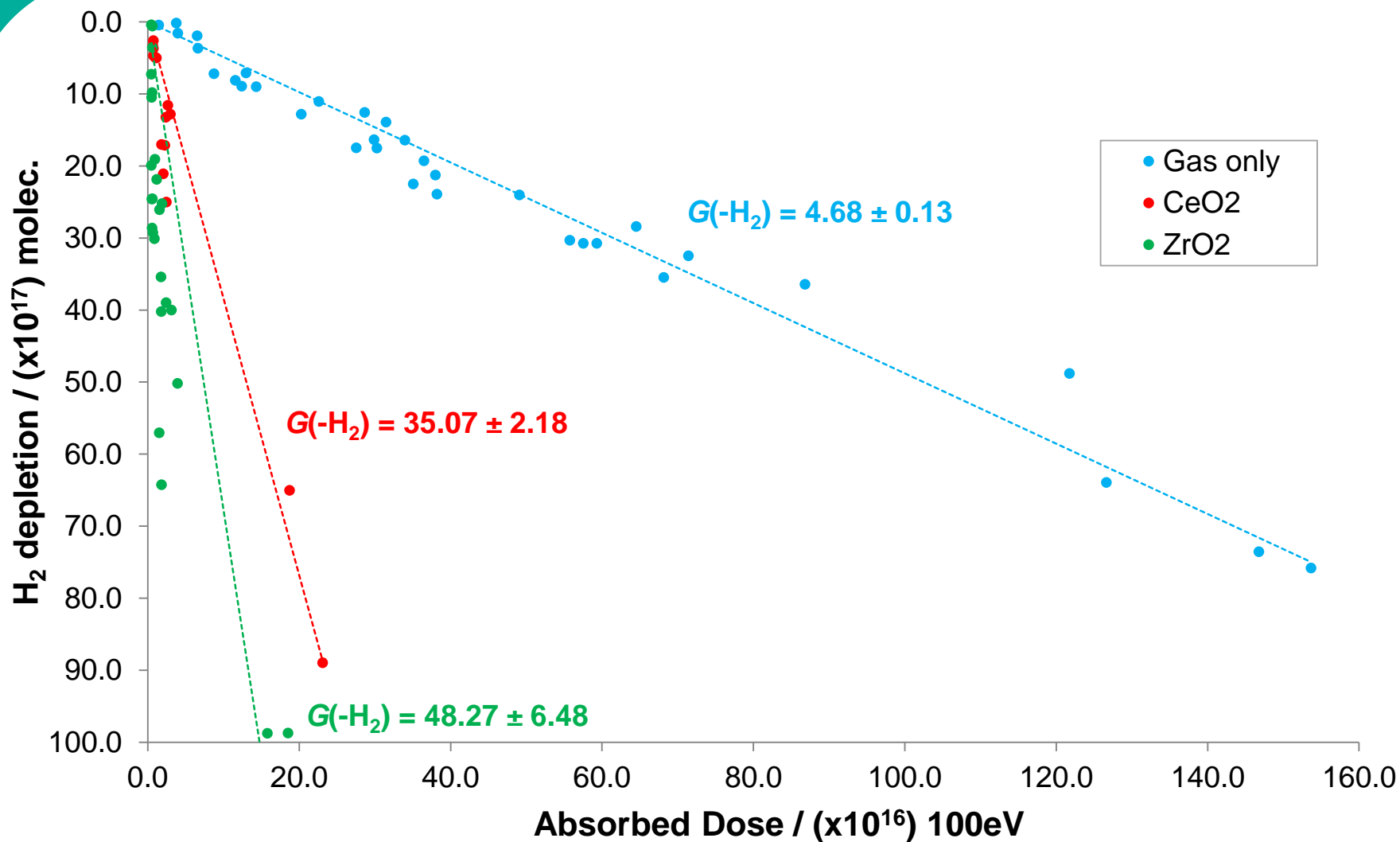
Results – H₂ depletion



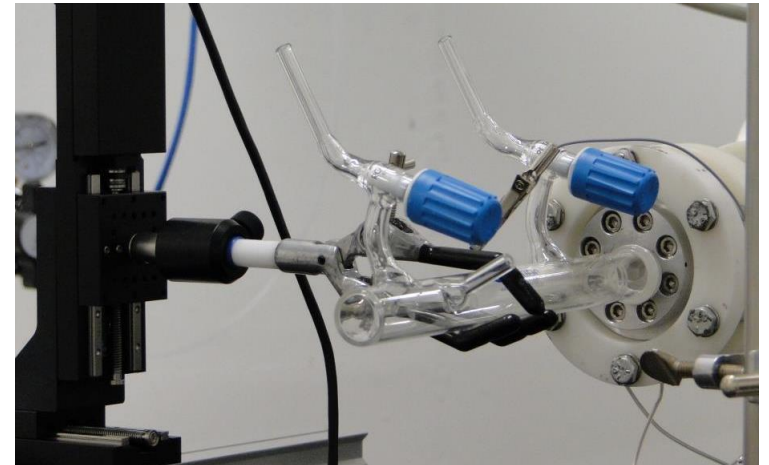
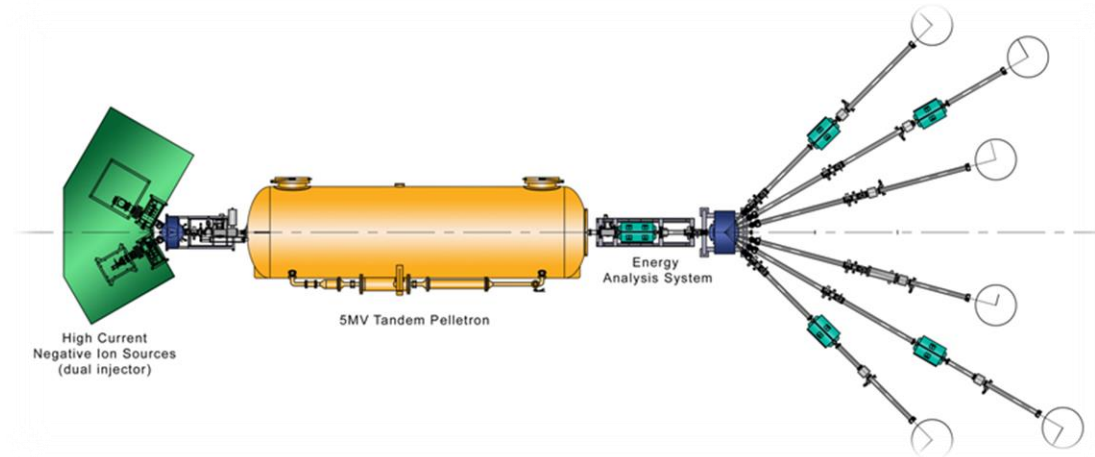
Results O₂ depletion



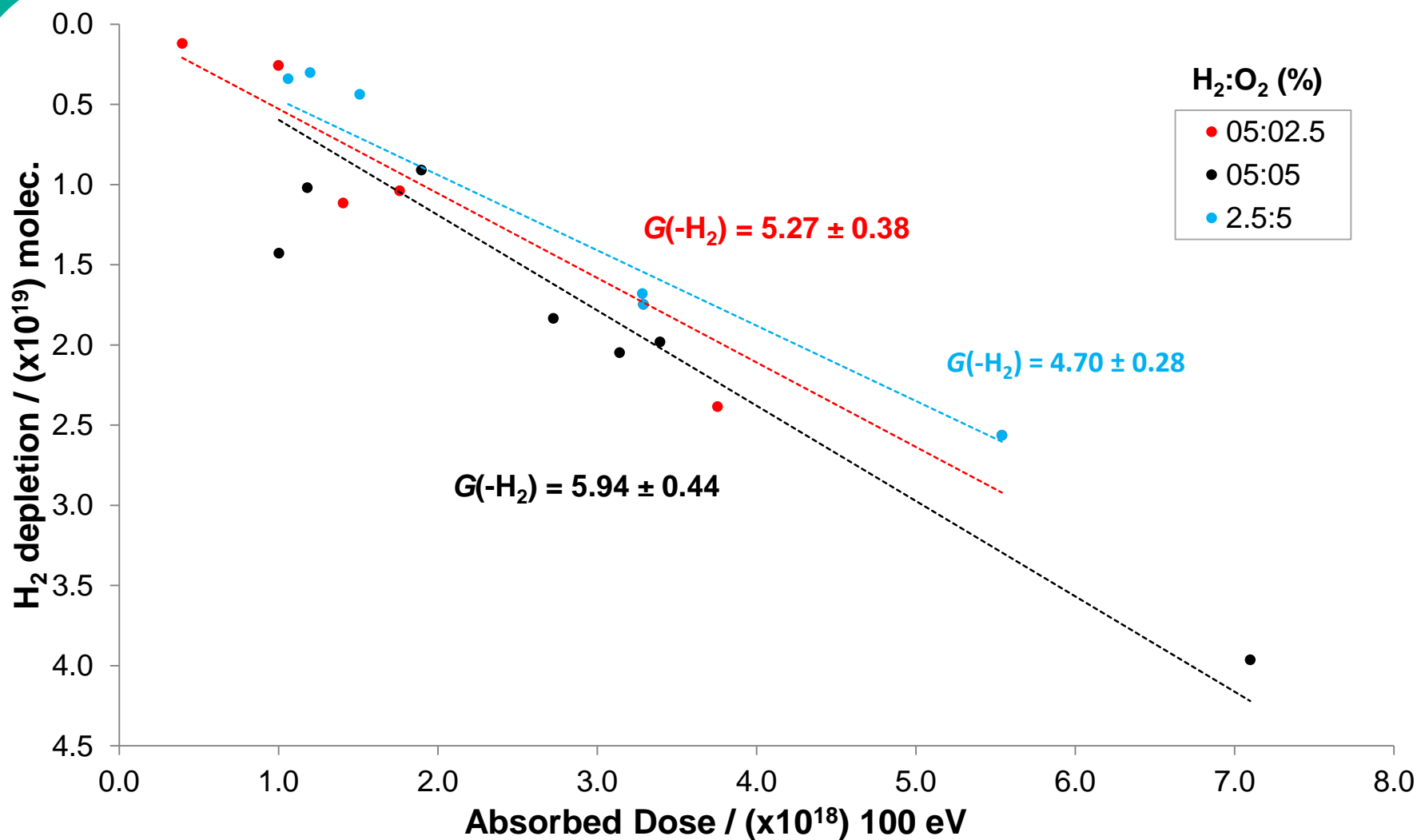
Results H₂ 5:5 (%)



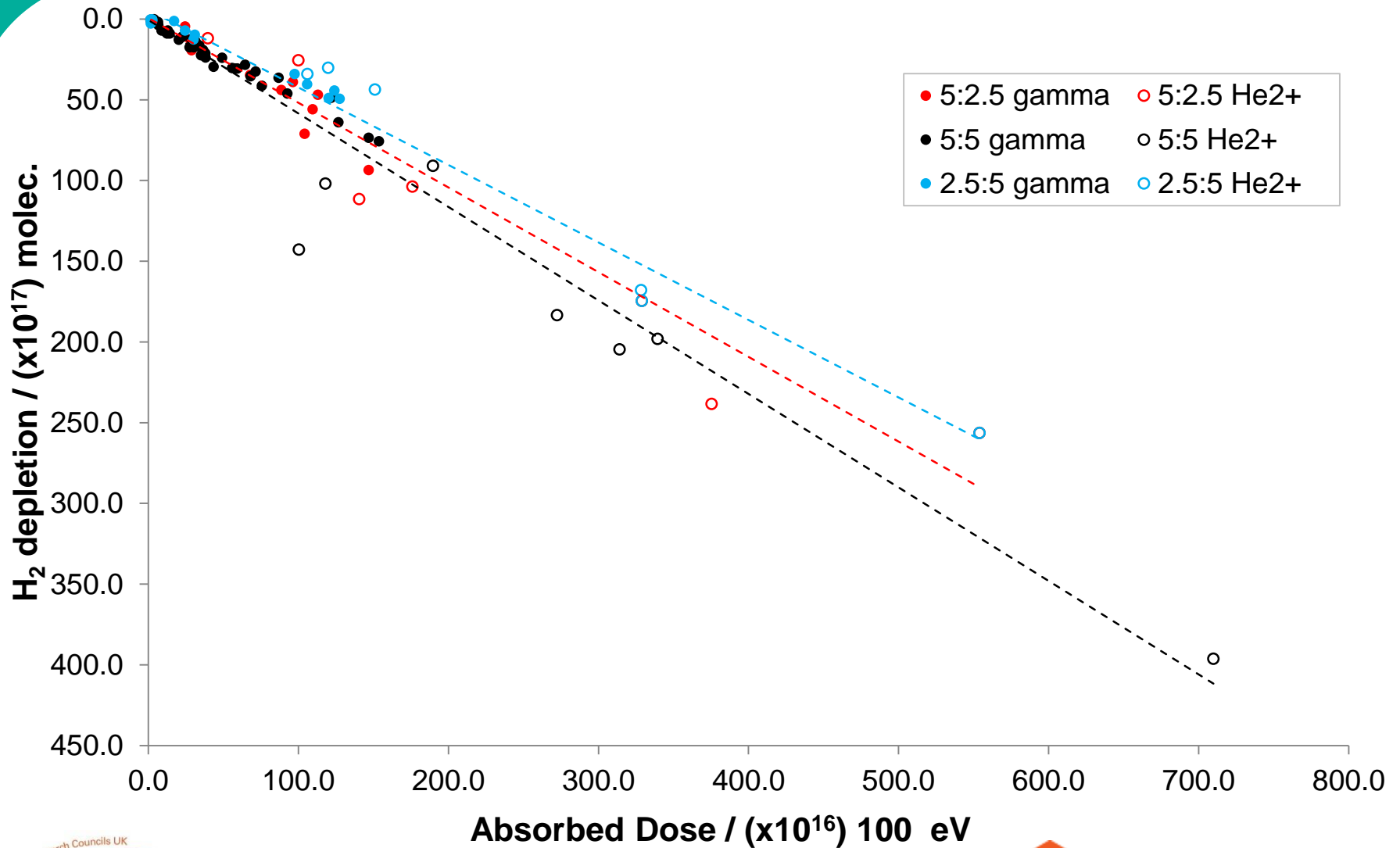
Experimental – Ion Source



Results H₂ – 5.5 MeV He²⁺ ions



LET comparison



Conclusions

- Rate of H₂ depletion is dependant on initial H₂ concentration
- Presence of an oxide surface increases depletion rate by an order of magnitude
- Depletion rate is independent of radiation type and intensity (no LET effect)
- Steady state not reached between recombination and radiolysis
- $2\text{H}_2 + \text{O}_2 \rightleftharpoons 2\text{H}_2\text{O}$

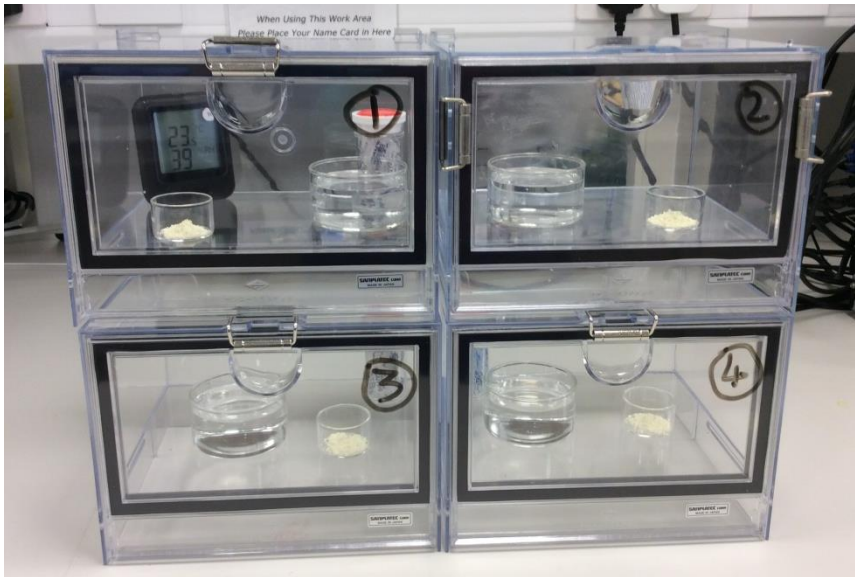
Future Work

- Heterogeneous experiments utilising ion accelerator
- Kinetic model of primary gas phase reactions
- Different oxide properties
- Better surrogate materials

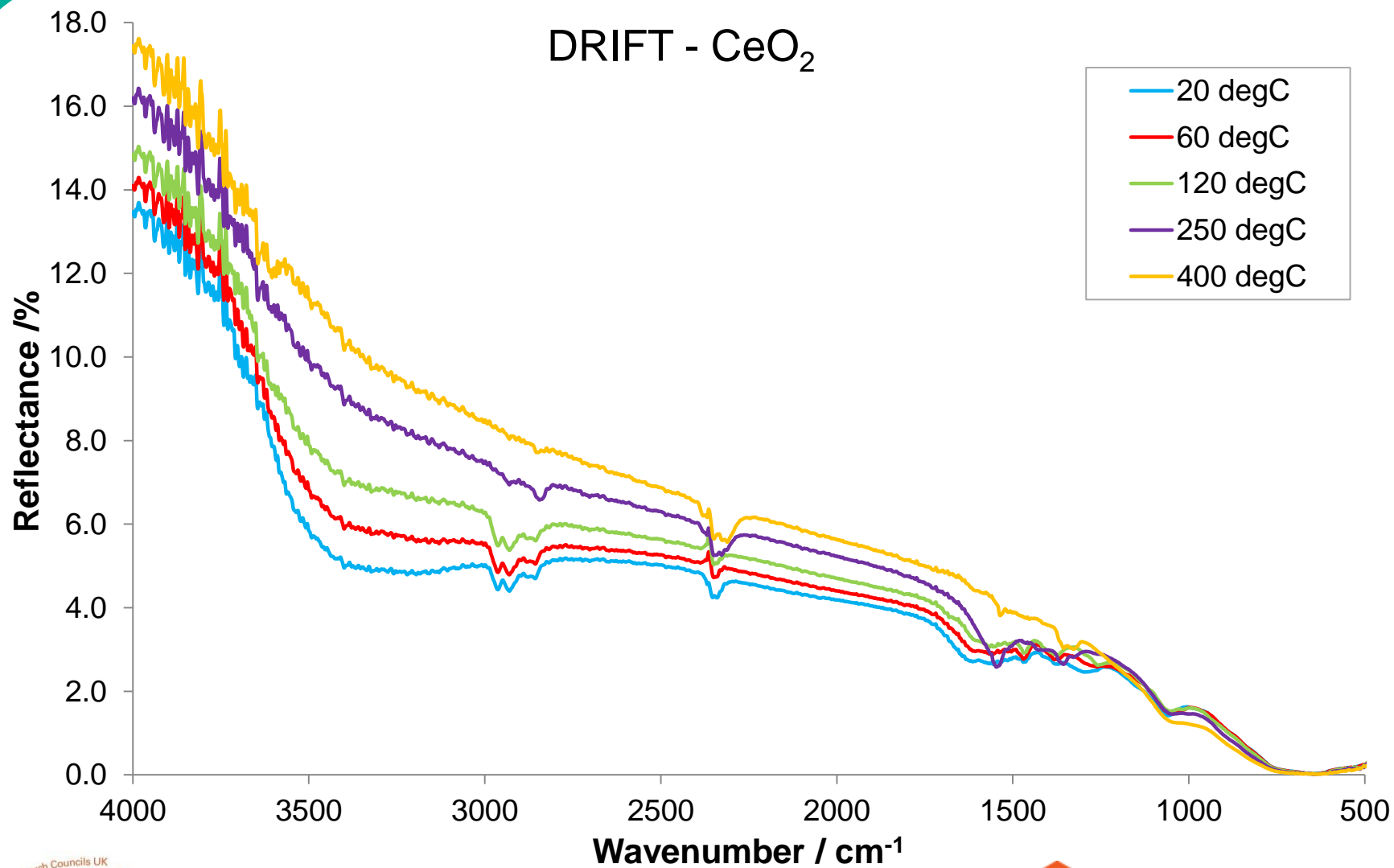
Further work cont.

- Investigation of water interaction with PuO_2
- Investigation of water adsorption with varying oxide properties
- Longer term sampling system to investigate H_2 production from water radiolysis adsorbed onto PuO_2

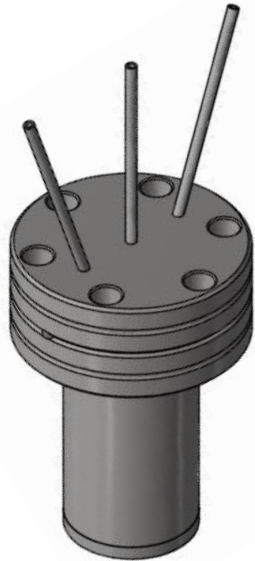
Water uptake on CeO_2



- CeO_2 baked and weighed pre-experiment
- %RH range 20-100
- Using saturated inorganic salt solutions



Vac. Sampling System



Acknowledgements

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