Bath Monash Global PhD Programme in Sustainable & Circular Technologies

Project Title:	Sustainable Water Splitting Using Electrocatalysts with Isolated Atomic Sites
Supervisors at Monash (Home institution)	Dr Cameron Bentley / Prof Stuart Batten
Supervisors at Bath (Host institution)	Dr Ulrich Hintermair / Prof Frank Marken
Indicative period at Bath	12 months

Project Summary (to include a brief description of the relevance to sustainable & circular technologies)

Water splitting by electrolysis is recognised to be one of the most promising approaches to convert and store renewable electricity in the form of hydrogen as a way to reduce our overreliance on non-renewable fossil fuels. To overcome high intrinsic energy barriers associated with this seemingly simple reaction, commercially viable water electrolysis requires the use of highly active and stable nanostructured electrocatalysts on both electrodes, such as porous films or nanoparticles with high surface-area-to-volume-ratios. Ideally however, all active atoms of the catalyst should be exposed to the reactants in solution and connected to the circuit to contribute to the catalysis as for example in natural enzymes. This is particularly important for rare/expensive precious metals, such as Pt, Pd, Ir, Ru, used for water electrolysis in acidic media. The ongoing search for low-cost, earth-abundant alternatives has spawned novel classes of material with well-defined, isolated active sites, for example single atom catalysts (SACs) and immobilised molecular catalysts (IMCs). While SACs and IMCs are optimal in terms of atomic utilisation, understanding catalytic mechanisms and the pathway(s) of catalyst deactivation on this scale is an ongoing challenge due to limitations in conventional instrumental approaches.

This PhD will introduce new paradigms for the fabrication and *operando* characterisation of SAC and IMC electrocatalyst materials by leveraging capabilities from Monash (electrochemistry, electrochemical imaging, SAC synthesis) and Bath (electrochemistry, spectroscopy, inorganic synthesis) and will:

- (i) Devise new methods to prepare highly-active, low-cost and ideally earth-abundant SACs and IMCs for use in water electrolysis;
- (ii) Use Monash and Bath-specialised, advanced (spectro)electrochemical characterisation tools with high spatiotemporal resolution to probe the active site(s) *in situ* during catalytic turnover, developing an in-depth understanding of catalytic activity/mechanisms;
- (iii) Perform complementary, advanced X-ray spectroscopy with the synchrotron light source to observe the potential- and/or time- dependent active site structure/composition in *operando*;
- (iv) Use the information from (i-iii) to engineer "next-generation" SACs/IMCs with enhanced function (*e.g.*, high activity, stability *etc.*), which will be tested under commercially-relevant conditions in a gas-diffusion electrode (GDE) setup.

The work aligns with sustainable and circular technologies since it aims to produce novel, low-cost, earth-abundant catalysts with isolated active sites that effectively minimizes/eliminates the need for rare precious metals in water electrolysis, addressing key research challenges spanning chemistry, material science and engineering. The work will contribute to Australia's National Hydrogen Strategy, which "*sets a vision for a clean, innovative, safe and competitive hydrogen industry that benefits all Australians. It aims to position our industry as a major player by 2030*" (https://www.industry.gov.au/data-and-publications/australias-national-hydrogen-strategy). Monash will provide expertise in SAC synthesis (Batten) and advanced electrochemical characterisation at the macroscale and nanoscale

(Bentley), while Bath will provide expertise in inorganic synthesis and NMR spectroscopy (Hintermair) and electrochemical and Raman spectroscopic characterisation (Marken). Once viable water electrolysis technologies are developed, the feasibility for industrial applications will be explored through collaboration with industrial partners, such as Woodside Energy through the Monash-Woodside partnership, taking advantage of well-established and readily available commercialization mechanisms.