Monash University, Low-cost, robust, high-activity water splitting electrodes R&D Project

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PROJECT SUMMARY AND SCOPE

Australia has immense renewable resources, the efficient harvesting of which will ensure future national energy security and support the development of a substantial export market for renewable energy. According to the Clean Energy Australia 2022 report, almost one third of national energy generation is already derived from renewable sources, and this contribution will only grow in the future.[1] To support these developments and future export of Australian renewables worldwide, electricity storage solutions that enable fast transport and long-term conservation on a terawatt scale are required. Among the options, renewable-powered electrolysis of water producing high-energy-density hydrogen fuel (H_2) presents one of the currently most feasible and sustainable solutions to this technological challenge.

Notwithstanding that the discovery of water electrolysis dates back more than two centuries[2] and intense recent initiatives from industry to implement the technology on a gigawatt scale,[3, 4] the price of hydrogen generated through this method still needs to be decreased to ensure competitiveness. The cost of electricity required for electrolysis is one major factor, although it depends on the installed capacity of the renewable solar, wind and tidal/current energy harvesting plants, which will expand in the coming years and thereby decrease the price substantially.[5, 6]

Another important contributor to the cost of electrolytic hydrogen are the electrolyser devices and their components, which need to be constructed using cheap and available materials to ensure the target terawatt scale of H_2 production is feasible.[7] The present project aims to address this impediment to the commercialisation of renewable-energy-powered electrolytic H_2 generation through the development of scalable methods for the fabrication of efficient, low-cost and robust electrodes for H_2 production from renewable energy sources *via* electrochemical water splitting.

Despite recent progress, most research on electrochemical water splitting has focused on catalyst discovery with the use of often prohibitively sophisticated fabrication methods. Contrasting this approach, the present project brings together complementary expertise of researchers from the Australian National University (ANU) and Monash University in the innovative fabrication of electromaterials to engineer efficient and cost-effective electrodes for efficient water splitting based on earth-abundant, low-cost elements such as iron. Of a specific focus to the project are highly-scalable electrode fabrication methods based on Flame Spray Pyrolysis (FSP) and electrodeposition. Other key features of the project include: design of efficient large-area water splitting electrodes, demonstration of the scalability of the developed fabrication methods and exhaustive stability tests under technologically relevant conditions.

High-performance electrodes are also indispensable for other electrolytic processes including renewable-powered electrosynthesis of ammonia from dinitrogen and water. Thus, the outcomes of the project will directly contribute to the development of innovative technologies for durable and cost-efficient H_2 and NH_3 production, resulting in immediate technological and commercial value.

Finally, through collaboration with AGL Energy, the present project was designed to assess the techno-economic prospects of the integration of electrolytic hydrogen in a "power-to-gas" mode into existing Australian gas supply networks. However, it is important to recognise that hydrogen is already an important commodity chemical, which is indispensable for several critical sectors of the chemical industry, like hydrocarbon refining and ammonia synthesis through a Haber-Bosch process. Moreover, H_2 is also expected to replace unsustainable reductants in other processes, like steel manufacturing. Hence, the scope of the techno-economic studies was expanded and refocused on a broader use of renewable hydrogen in Australian industry, specifically as a (i) feedstock synthesis for ammonia production in Queensland & Pilbara, (ii) pulverised coal replacement for green(er) steel at Whyalla Steelworks, and (iii) fuel for transport and logistics in Melbourne Metropolitan Distribution.

DIFFICULTIES EXPERIENCED

While the restrictions due to the worldwide spread of the COVID-19 virus have affected the progress of our research substantially through the periodic lock-downs of the laboratories and severely limited access to several key research facilities, timely establishment of the alternative work arrangements decreased the impact of the global pandemic situation on the progress of this project. Both Monash and ANU research teams have successfully achieved the goals identified in the research agreement.

Another key difficulty experienced was in the engagement with the traditionally conservative Australian industry, who have become even more cautious and resistive to active engagement with academia under the current circumstances of the aggravating economic challenges worldwide. Notwithstanding these challenges, Chief Investigators (CIs) have made a significant progress in establishing new productive collaborations with industry in the field of this technology.

KEY HIGHLIGHTS AND ACHIEVEMENTS

All planned outcomes and milestones of the project, which are summarised below, have been successfully achieved.

Major outcomes of the project:

1 Progression of the flame spray pyrolysis water splitting electrode fabrication technology to Technology Readiness Level (TRL) 5;
2 Creation of a techno-economic model and its application for the analysis of the use of sustainable hydrogen in different sectors of the Australian industry;
3 Expanded connections to Australian and international industrial partners in renewable hydrogen sector for commercialisation of the developed technologies, and exploration of pathways for export of renewable Australian hydrogen; and
4 Greater awareness in the public, government and industry sectors around the development of renewable hydrogen technology in Australia.

Final milestones achieved upon the completion of the project:

Milestone 3.1: Demonstration of a laboratory roll-to-roll flame spray pyrolysis system for the fabrication of electrodes with area ≥10 cm^2.

Milestone 3.2: Demonstration of a water splitting system operating at efficiency ≥77% and showing degradation ≤10% over 2 weeks at 80°C.

Milestone 3.3: Demonstration of hydrogen generation in an electrolyser prototype at ≥0.50 L h^{-1}.

Milestone 3.4: Techno-economic model of the use of sustainable hydrogen in different sectors of the Australian industry.

In what follows, selected achievements of the project are highlighted.

Roll-to-roll flame-spray-pyrolysis catalyst fabrication facility

We have successfully engineered a roll-to-roll flame spray pyrolysis (FSP) setup at ANU. As shown in **Figure 1**, the set-up includes two main parts: the roll-to-roll and the flame spray pyrolysis systems, both of which can be easily and individually adjusted to match the fabrication requirements. For example, one can tune the speed and direction of the rolls so that it can run faster or slower to control the catalyst loading. The width of the substrate/sheet/belt can reach ~12 cm, but can be expanded through the installation of additional FSP nozzles. The length of the rolled substrate in our current setup can be as long as 10 m, but can be even longer as it is only limited by the size of the rolls used. The distance between the flame nozzle and the rolling substrate is adjustable, which enables facile control over the deposition temperature and thereby the catalyst composition, structure and morphology.

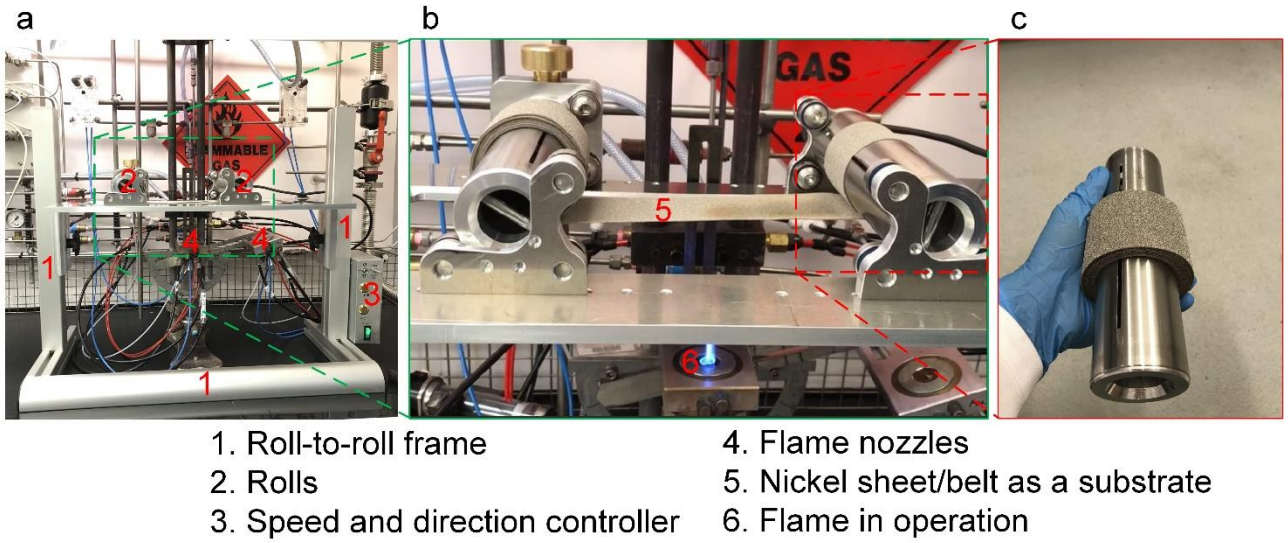

Figure 1. Photographs of the roll-to-roll FSP electrode fabrication facility: (a) general overview, (b) setup in operation, and (c) a roll of nickel foam sheet.

The FSP part of the system is outstandingly versatile in terms of the chemical composition and physical characteristics of the deposited materials and substrates. Our flame synthesis setup was used to produce a wide variety of materials such as FeO_x, Co_3O_4, SnO_2, MnO_x, ZnO, $BiVO_4$, Bi_2O_3, Au-Bi_2O_3, $BiFeO_x$, SiO_2, Au, WO_3, to name a few.[8-25] As a substrate, the FSP can use either flat supports, *e.g.* metal foils, or more advanced high-surface area materials like grids, meshes, fibre cloths and foams. Moreover, the properties of the deposited materials can be easily adjusted by altering the spray and pyrolysis conditions.

As a demonstration of the capabilities of the new facility, we have deposited a high-performance Co_3O_4 electrocatalyst on a nickel foam in a roll-to-roll mode as shown in **Figure 2a-b**. The electrodes with an area of >200 cm^2 were obtained within 10 minutes. Visual inspection and microscopic characterisation attest to the homogeneous distribution of a thin layer of Co_3O_4 over the large surface area (**Figure 2c-d**). Such flame-made Co_3O_4 materials exhibit excellent water electrooxidation activity under alkaline conditions, reaching remarkable mass-weighted performance reaching 2000 A g^{-1} (ampere per gram of the catalyst) at an overpotential of 0.4 V.[26]

To advance the technology even further, the CIs on the present project have been successful in securing extended funding through the Australian Research Council (ARC) Linkage Infrastructure, Equipment and Facilitates (LIEF) scheme for the development of an innovative flame-based facility for the production of non-oxide based electromaterials. This advanced strategy will enable one-step fabrication of nanostructured coatings of metal pnictides and chalcogenides in a single, very fast step avoiding the need for the traditional high-temperature treatment in a furnace. Clearly, the new

FSP setup is directly compatible with the versatile roll-to-roll system developed in the present project. This new facility is currently in production and will be installed in Prof Tricoli's Sydney laboratories, providing novel capabilities for Australian researchers for creation of functional electromaterials and electrodes for green hydrogen production and other sustainable technologies.

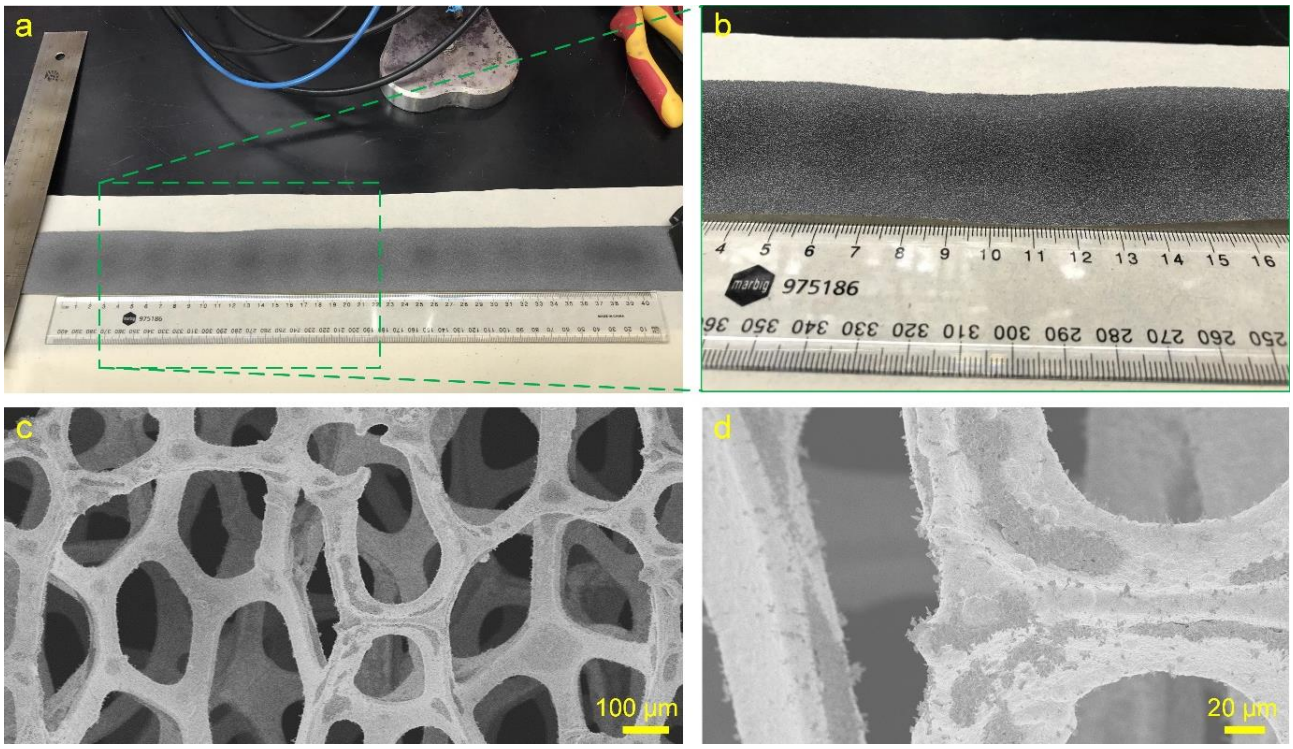

Figure 2. (a-b) Photographs and (c-d) scanning electron micrographs of a >200 cm^2 Ni foam substrate modified with a Co_3O_4 catalyst using roll-to-roll set-up shown in **Figure 1**.

High-performance water electrolysis under alkaline conditions

In the course of the project, we have introduced FSP methods for the fabrication of a range of water splitting electrodes with various catalysts, majorly based on the first row transition metals. At the same time, aiming to achieve our initial goal of developing materials based on truly abundant and cheap elements, we have focused our major efforts on catalysts containing iron as the major active element. In 2021, the amount of iron ore mined was 2.6 billion tonnes, vastly exceeding the overall amount of all other metals mined together over the same period [USGS Mineral Commodity Summaries, 2022]. Thus, the water splitting electrodes highlighted in the present report are based on a cheap iron catalyst deposited as thin nanostructured coatings on various substrates.

To demonstrate scalability of our fabrication strategy, we used 176 cm^2 circular nickel foam substrates (**Figure 3a**) to deposit an iron-based catalyst in the form of high-surface-area interconnected network of highly-dispersed 20-30 nm particles (**Figure 3b**).

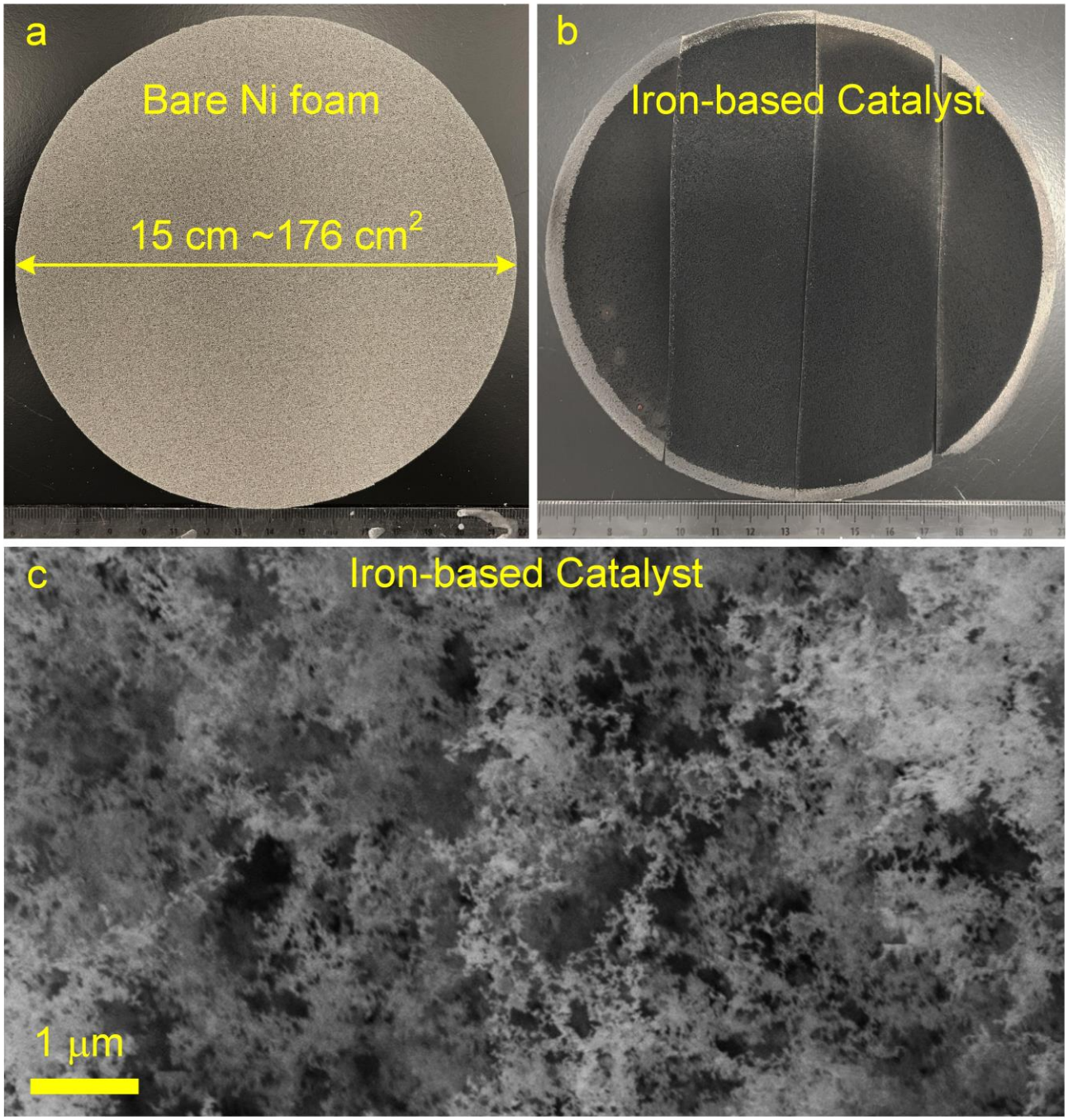

Figure 3. (a) Photographs and (b) scanning electron micrographs of a high-area Ni foam substrate modified with an iron-based catalyst made using the flame pyrolysis in CI's laboratories.

The use of such electrodes as both cathodes and anodes for water electrolysis in alkaline electrolyte solutions demonstrates their competitive catalytic performance, especially for the more kinetically challenging anodic oxygen evolution reaction (**Figure 4a**). Specifically, the electrodes required the application of resistance-free potentials of just 1.47 ± 0.02 V and -0.28 ± 0.02 V *vs.* reversible hydrogen electrode (RHE) to sustain the oxygen and hydrogen evolution reaction at the rates of 200 mA cm^{-2} at 80 ± 1 °C, respectively. These FSP-made electrodes were also tested as both anode and cathode for water electrolysis in regular two-compartment laboratory cells separated with an

AGFA "Zirfon" commercial H_2 separator filled with 1 M (mol per litre) KOH. The overall cell potential required for water electrolysis at 200 mA cm^{-2} and 80 ± 1 °C was 1.76 ± 0.02 V, which corresponds to the thermoneutral efficiency of 84 ± 1 %.

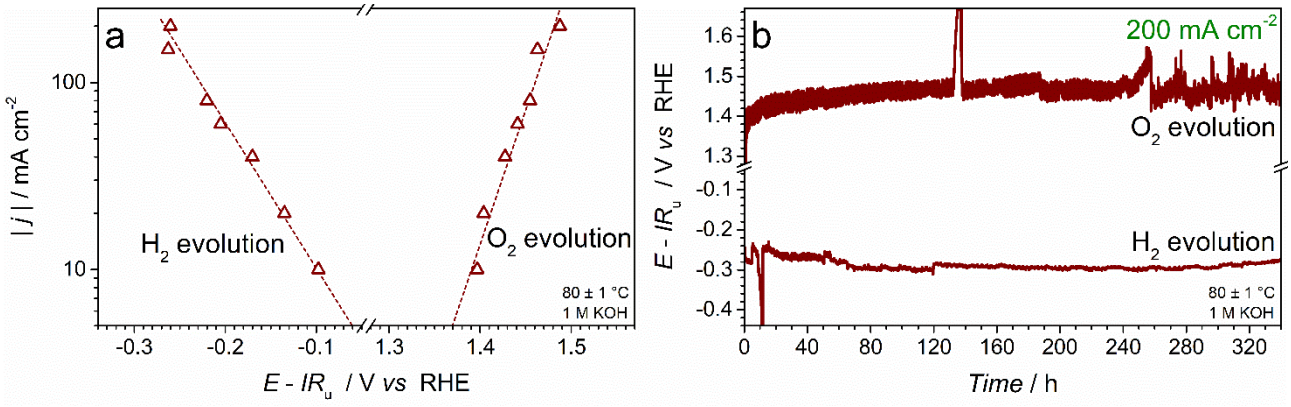

Figure 4. Electrocatalytic performance of the Ni-foam electrodes FSP-modified with an iron-based catalyst. (a) Polarisation plots, and (b) 2 week performance tests at a constant current density of 200 mA cm^{-2} for the hydrogen and oxygen evolution reactions in 1 M KOH at 80 ± 1 °C. The electrolyte solution was saturated (1 bar) with H_2 and O_2 during the hydrogen and oxygen evolution tests, respectively. All data were corrected for ohmic losses (uncompensated resistance, R_u = 1.00 ± 0.02; the size of the electrodes was 0.5 cm × 0.5 cm = 0.25 cm^2).

Equally importantly, tests at an applied current density of 200 mA cm^{-2} demonstrated outstanding durability of the electrodes developed in the present project on a time-scale of two weeks (**Figure 4b**). Upon stabilisation over the initial day of operation corresponding to approximately 2 % loss in the efficiency (approximately 0.04 V), the performance remains stable for the remaining period of test, *i.e.* demonstrates 0% degradation.

The performance of the Ni-foam electrodes FSP-modified with an iron-based catalyst was also tested in an electrolyser prototype constructed for the purposes of the present project (**Figure 5a**). The identical cathode and anode with an active area of 5 cm^2 each were positioned as close as possible to different sides of a commercial Zirfon separator. As electrolyte, 6 M KOH aqueous solution was used (saturated with H_2 at 1 bar on the cathode side to avoid unproductive O_2 reduction reaction), and the tests were run at 80 ± 1 °C.

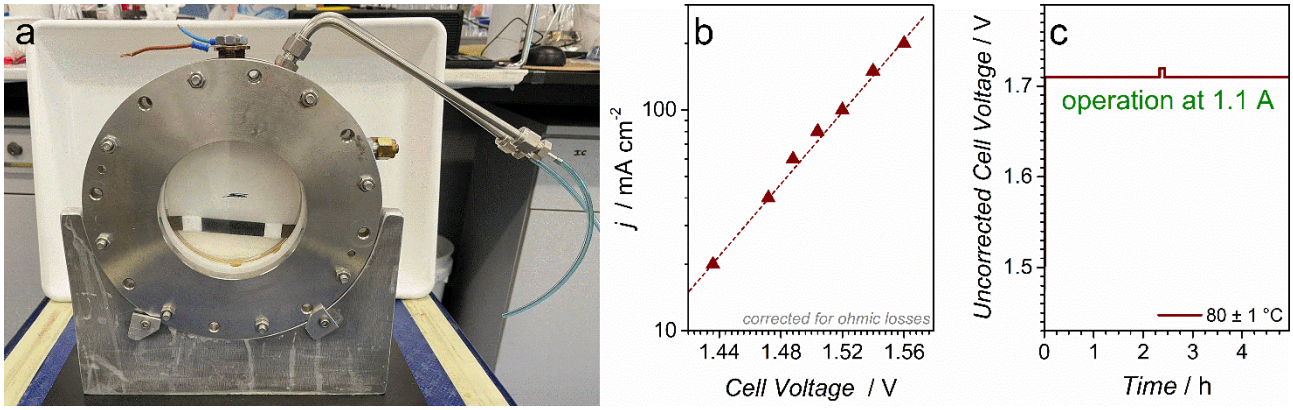

Figure 5. High current water electrolysis with the Ni-foam electrodes FSP-modified with an iron-based catalyst. (a) Photograph of the electrolyser setup, (b) polarisation plots, and (c) operation of the electrolyser at 1.1 A corresponding to a current density of 0.22 A cm^{-2} and 0.50 L h^{-1} hydrogen production rate. Test were undertaken at 80 ± 1 °C using 6 M KOH which was additionally saturated with H_2 (1 bar) on the cathode side. Data in panel b was corrected for ohmic losses, while panel c shows raw, uncorrected cell voltage. The size of the electrodes was 1 cm × 5 cm = 5 cm^2.

The potential dependence of the logarithm of water electrolysis current corrected for the resistance losses was close to linear, attesting to minimal effects of mass-transport on the performance within the FSP-synthesised electrodes up to 220 mA cm^{-2} current density (**Figure 5b**). This is a highly favourable observation of a practical significance. The target current of 1.1 A, corresponding to the H_2 generation rate of 0.50 L h^{-1} at 80 ± 1 °C was successfully achieved at the overall non-corrected potential of the electrolyser of 1.71 ± 0.01 V (**Figure 5c**), which corresponds to the thermoneutral energy efficiency of water electrolysis of 87 ± 1 %.

Iridium-free systems for water electrolysis under acidic conditions

Broadening our scope for both facile and scalable electrode fabrication capabilities of earth-abundant water electrolysis catalysts, electrodeposition of catalytic materials became a second core focus in the development of anode materials that can withstand the oxygen evolution reaction at low-pH, elevated temperatures and elevated current densities. Electrodeposition is a highly attractive approach for large-scale electrode fabrication, as its industrial maturity and tunability provides robust and highly consistent procedures for the production of low-cost electrolytic materials on an array of industrially usable electrode substrates and geometries.

This concept formed the basis of the development for a highly stable cobalt- and iron-doped lead(IV) oxide electromaterial. The catalytically active cobalt-iron species are incorporated and stabilised by the formation of the conductive and corrosion resistant β-PbO_2 matrix that is formed at pH ≤ 1, high current densities ≥ 0.5 A·cm^{-2} and at temperatures as high as 80 °C.[27, 28] This formative work

supported further investigation and development of anode materials, awarding high-performance and earth-abundant oxygen evolution reaction catalysts that are capable of withstanding the harsh acidic conditions owing to the stabilising effects of the new bismuth[29, 30] and antimony oxide[31] stabilising matrices.

Of the developed materials, the cobalt-iron-lead oxide anode catalyst was selected for extended testing based on its remarkable ability to uphold stable water oxidation currents in pH ≤ 1 and at 80 °C. At the initial stage of the project, we demonstrated high-performance operation of the [Co-Fe-Pb]O_x anodes in the presence of dissolved precursors for all three components of the catalyst,[28] which would not allow its integration into a complete electrolyser setup due to the poisoning effect of lead on the cathode activity. An important further advancement was the development of a "cobalt-selective" self-healing mode of operation of [Co-Fe-Pb]O_x in the presence of only dilute amounts of dissolved cobalt precursor species (less than 5 mM), which provides stable anode operation without poisoning of the H_2 evolving cathode.[27]

This was demonstrated during a two-week water electrolysis test with a model "beaker"-type cell filled with aqueous 0.5 M H_2SO_4 electrolyte solution containing 5 mM Co^{2+}. Stable operation in a two-electrode configuration a current density of 100 mA cm^{-2} and a temperature of 80 ± 1 °C required a stabilised cell voltage of 1.84 ± 0.01 V corresponding to a thermoneutral energy efficiency of 80 ± 1 % **(Figure 6)**. This performance is one of the highest reported so far for water electrolysis under acidic conditions with a noble-metal free anode. Stabilisation during the initial several days of operation induced only a minor increase in the cell potential by *ca* 0.04 V (corresponding to approximately 2 % loss in efficiency) and then remained stable for the rest of the 2-week-long test.

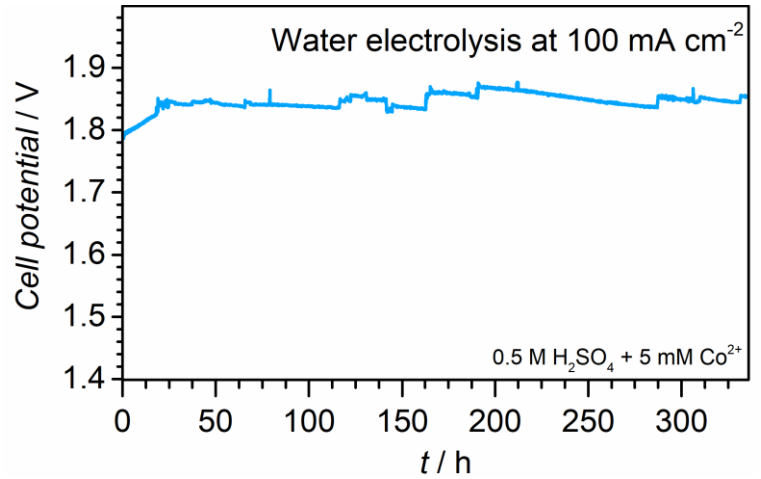

Figure 6. Galvanostatic water electrolysis at 100 mA cm^{-2} and 80 ± 1 °C in the presence of 0.5 M H_2SO_4 electrolyte and 5 mM Co^{2+} using PtTi-felt electrodes functionalised with [Co-Fe-Pb]O_x (anode) and 0.1 mg cm^{-2} 20 wt.% Pt/C catalyst ink (cathode). [Co-Fe-Pb]O_x catalyst film was prepared by 2 h galvanostatic oxidation (100 mA cm^{-2}) of 0.5 M H_2SO_4 solutions containing 5 mM Co^{2+}, 1 mM Fe^{3+} and 0.5 mM Pb^{2+} at 80 ± 1 °C.

Considering the requirement of the presence of the Co^{2+} precursor for stable operation of the electrodeposited [Co-Fe-Pb]O_x anode catalyst developed in the project, we have designed an electrolyser that would be able to operate under such conditions **(Figure 7)**.

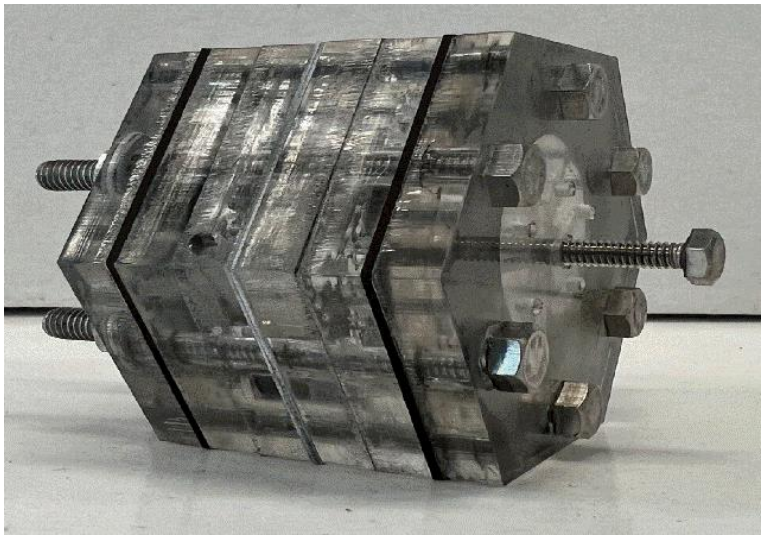

Figure 7. Photograph of an electrolyser prototype designed for the operation of a [Co-Fe-Pb]O_x and similar noble-metal-free anode catalysts with a geometric surface area of the electrodes of 7 cm^2.

Using this prototype, we have demonstrated water electrolysis at an overall current of 1.1 A at the temperature of 80 ± 1 °C using the [Co-Fe-Pb]O_x anode. These results have formed a basis for our new research project focusing on the optimisation of the electrolyser architecture and electrode composition, which is currently ongoing and is expected to eventuate in the demonstration of an inexpensive and robust acidic water electrolyser at a >100 cm^2 electrode size scale.

Techno-economic model and commercialisation prospects

One of the features of the present ARENA-funded project is the collaboration between Monash researchers and the AGL Power Development team on the techno-economic analysis of the commercialisation prospects of the hydrogen derived through water electrolysis powered by renewable electricity. This study was supported by extensive discussions across AGL and associated companies on a range of key topics, including: Wholesale Electricity Markets, Physical Markets Trading in the NEM, AGL Futures Portfolio, Levelized Cost of Hydrogen, Hydrogen Energy Supply Chain Project (HESC), Valuations & Uncertainties in Hydrogen Projects, Technical Considerations of Hydrogen (Altona North Electrolyser) and the Western Sydney Gas Project (WSGP). A financial model resulting from the Monash-AGL collaboration was used for the analysis of three selected cases for the use of green hydrogen in Australian industry.

One key outcome of this analysis was the identification of the cost of the water electrolysis devices, which is to a significant extent affected by the price of the electrodes, as a more significant parameter than the energy efficiency of electrolysers in terms of the cost-efficiency. This means that development and implementation of electrodes that might be not as efficient as the currently used noble-metal based systems, but are orders of magnitude less expensive, will substantially improve the cost-effectiveness of the production of H_2 by renewable-powered water electrolysis. This outcome aligns with the practical outcomes of the project in the design of catalysts based on inexpensive earth-abundant metals, like iron.

Apart from the components of the catalyst, another key contributor to their cost is the method of their production, which should be cheap, fast and scalable to satisfy the requirements of the terawatt-scale H_2 generation. The roll-to-roll flame spray pyrolysis method developed herein perfectly matches all of these requirements, and presents an industry-ready process. Flame synthesis is currently utilised for the commercial production of nanoparticle commodities such as carbon black, fumed silica and P25 photocatalysts at a scale of megatons per year. However, translating the flame route fabrication benefit to the fabrication of electrocatalysts for the hydrogen production has been challenging due to the subtle nature of the effects of deposition conditions on the catalytic activity. Furthermore, fabricating thin catalytic layers of flame-made nanoparticles onto electrolyser electrodes by standard approaches such as drop-casting and screen-printing usually leads to imperfect morphology, where a significant fraction of the catalyst mass is unutilised. The roll-to-roll direct deposition technology, developed here, combines the scalability and low-cost production of materials by flame (e.g. fumed silica current price is US$2500/Ton) with the capability to finely control the resulting catalyst film morphology, mass and porosity, leading to their efficient utilisation. In average, a thickness of less than 200 nm of catalysts is required to coat the electrode resulting in *ca* 0.6 g m^{-2}, and thus a material cost of less 0.1 US$ m^{-2} for preparing electrodes for highly energy-efficient (87% demonstrated herein) water splitting into hydrogen, assuming a 50% yield of deposition and similar price as fumed silica when the flame process is used in large scale.

The above conclusions are already being confirmed through the rapidly expanding connections of the Monash-ANU teams with industry partners developing sustainable water electrolysis technologies. Of a particular note is the new project funded by Woodside Energy at Monash focused on the development of efficient electrodes and devices for the electrolytic hydrogen generation from sea water. Other connections of ANU and Monash researchers to other major Australian and German companies are currently at the negotiations stage. These developments have emerged from the outcomes of the ARENA-funded project.

SUMMARY OF KNOWLEDGE SHARING ACTIVITIES

The outcomes of the ARENA-funded project and our broader research in the renewable energy technologies are regularly disseminated through various mechanisms at different levels.

Of a particular note are the following activities in this domain:

(1) Engagement with academia, industry and government

- Dr Simonov will participate in a delegation visit to Japan (Dec 2022) aiming to bring together researchers from Japanese and Australian industry and research institutions to explore R&D opportunities that will accelerate the development of the collaborative Australia-Japan hydrogen industry.
- Woodside Energy Ltd – a leading Australian energy company, is funding a new research project (2021-2024) led by Dr Simonov and Prof MacFarlane on the development of seawater electrolysis technologies [https://www.monash.edu/woodside/energy-partnership/research-themes]. This new project was a direct extension of the present ARENA-funded activity.
  The Woodside project was preceded by a scoping feasibility study in 2020 funded by Woodside. The establishment of this connection and current operation of the project include extensive discussions over a significant number of visits of Monash researchers to Woodside headquarters in Perth and Woodside top-managers to Monash Clayton campus as well as video/phone conferencing.
- Prof Tricoli, Prof Macfarlane and Dr Simonov have secured funding from the Australian Research Council (ARC) through the Linkage Infrastructure, Equipment and Facilities (LIEF) program to a develop an innovative flame-based facility for the production of non-oxide based electromaterials. This facility is currently in production and will be installed in Prof Tricoli's Sydney laboratories, providing novel capabilities for Australian researchers working in electrochemistry of green fuels.
- Dr Simonov has been awarded an ARC Future Fellowship on the development of innovative electrocatalytic technologies for energy storage and transformation. The project involves active interaction with Australian and German industry partners in sustainable production of hydrogen, ammonia and fertilisers.
- Prof Tricoli has been awarded an ARC Future Fellowship on the development of porous electromaterials for electrolysers, batteries and electrocatalytic systems for green fuel production. The project involves active interaction with Australian, Italian and USA industry partners in the broad field of electromaterials.
- Through an award of a Fulbright Fellowship by the American-Australian commissions, Prof Tricoli spent 3 months in the group of Prof J. Li at the Massachusetts Institute of Technology (Boston, USA), developing collaborative projects for the scalable design and high-throughput search of electrocatalysts for water splitting.
- Dr Simonov and Prof MacFarlane have initiated a new collaborative project with Energys Australia Pty Ltd – one of the leading Australian start-up companies in the green hydrogen domain.

- Prof Tricoli, Dr Simonov and Prof MacFarlane have established major connections with ENI – an Italian multinational energy company – *via* Australian and Italian head-office through several meetings, where results from the current ARENA-funded project were presented.
- Prof Tricoli has negotiated future collaborations in the water electrolysis and electrode fabrication development with the Italian Institute for Technology (IIT).
- In collaboration with academic colleagues and industry partners throughout Australia, Prof Tricoli, Dr Simonov and Prof MacFarlane are currently working on a proposal for a Cooperative Research Centre (CRC) for Scaling Green Hydrogen. The submission is planned for the next round in early 2023.
- Dr Simonov and Prof MacFarlane held a virtual meeting with the delegation from IXOM – a leading company in Australia and New Zealand in industrial electrolysis, and have hosted an IXOM representative at Monash University. The discussions focused on a potential collaborative Monash-IXOM PhD project on further development of the electrolyser technology introduced in the present ARENA-funded project.
- Dr Simonov acted as a consultant for an Australian company FutureBus Pty Ltd on the development of hydrogen technologies in Australia. FutureBus became aware of the Monash expertise in the renewable hydrogen area through the information on the present ARENA project on the ARENA website.
- Dr Simonov and Prof MacFarlane have been involved as CIs in the German based PrometH_2eus project (part of the H2GIGA initiative) of the German Federal Ministry of Education and Research (BMBF). This project brings together 12 leading German universities, industry giants like thyssenkrupp AG and De Nora, and the only international partner being the Monash group of the CIs on this ARENA-funded project. The project aims to advance the technology of alkaline water splitting for scalable H_2 generation.
- Dr Simonov and Prof MacFarlane have established connections to an Australian company eWater systems Pty Ltd who distribute the disinfection units operating through electrolysis of saline water.
- Dr Simonov has been involved as a Chief Investigator (CI) in a GlobH2 ARC Industrial Transformation Training Centre for The Global Hydrogen Economy led by UNSW (2021-2026). The centre aims to transform Australia into a hydrogen powerhouse by building enabling capacity in hydrogen innovation in a short timeframe. The Centre aims to generate new technologies and equip a future workforce of industry-focused engineers with advanced skills for development and scaling-up of hydrogen generation and transport.
- Through a DAAD (German Academic Exchange Service) – UA (Universities Australia) exchange grant, Monash researchers led by Dr Simonov undertook collaborative research with scientists from Max Plank Institute for Chemical Energy Conversion (Mulheim, Germany) on the catalysts developed in the present ARENA-funded project.
- Monash CIs have hosted several high-level delegations to our laboratories:
  - a delegation from Germany led by the German National Academy of Science and Engineering (acatech) and the Federation of German Industries (BDI) focussed on Energy Futures based on hydrogen and ammonia (Sep 2019);

- a delegation headed by Mr H. Kajiyama, Japanese Minister of Economy, Trade and Industry, along with the Australian Minister for Trade, Tourism and Investment, Senator S. Birmingham (Jan 2020);
- a delegation headed by the President of the National Academy of Engineering Korea Prof O.-K. Kwon (Mar 2020).

All visits included a showcase of the Monash & ANU expertise in the renewable hydrogen and ammonia technologies, as well as discussions of the joint development of the renewable hydrogen and ammonia technologies between Australia and Japan, Korea and Germany.

(2) Media highlights

Our work on the development of new low-cost, robust water splitting electrodes and devices has been broadly highlighted in various media:

- RSC Chemistry World [https://www.chemistryworld.com/news/uk-project-aims-to-cut-the-cost-of-producing-clean-green-hydrogen/4011788.article#/]
- Monash.Lens [https://lens.monash.edu/@science/2019/09/24/1376698/electrolysis-breakthrough-could-solve-the-hydrogen-conundrum]
- Nature Blog [https://chemistrycommunity.nature.com/users/253869-alexandr-simonov/posts/48277-intrinsically-stable-in-situ-generated-electrocatalyst-for-long-term-oxidation-of-acidic-water-at-up-to-80-c]
- Monash University News [https://www.monash.edu/science/news/current/monash-leads-the-way-in-a-green-chemistry-breakthrough-for-renewables/_nocache]
- PV-Magazine [https://www.pv-magazine-australia.com/2019/05/16/monash-researchers-achieve-green-hydrogen-breakthrough/]
- Renew Economy [https://reneweconomy.com.au/scientists-edge-closer-to-stable-cheap-green-hydrogen-production-78478/]
- Sustainability Matters [https://www.sustainabilitymatters.net.au/content/sustainability/case-study/researchers-closer-to-affordable-green-hydrogen-1543192365]
- National Resources Review [https://www.nationalresourcesreview.com.au/news_article/australian-researchers-lead-the-way-in-a-green-chemistry-breakthrough-for-renewables/]

Prof MacFarlane was featured in the Channel 9 story on the electrolytic hygiene systems developed by eWater systems P/L: https://www.youtube.com/watch?v=WbXR-7fPH3Y

(3) Public presentations

- Dr Simonov and Prof MacFarlane participated as panellists for a public conference *Green Hydrogen for a Sustainable European Future*, Berlin Germany, May 2022.
- Presentation of the experimental "Solar Fuels" rig constructed through the present ARENA-funded project to the public visitors during the Monash Open days (2018-2022).

- Highlights of the green energy technologies during the Science Industry Nights at Monash University (2020-2022)
- PhD candidate supervised by Dr Simonov, presented and advertised the renewable energy concepts and science in general for the International Women's Day (in Science) at Melbourne Girl's College.
- Youtube video highlighting the *operando* analysis of electrocatalysts [https://www.youtube.com/watch?v=Z_UYpZC8xu0]

(4) Conference presentations

ANU and Monash researchers have presented the results of the present project at a number of major conferences. Note that the technologies for water electrolysis developed herein directly support sustainable synthesis of ammonia, which is also a major research theme in our groups. Many of our presentations addressed both technologies in an integrated way.

- **Sustainable Technologies for the Synthesis of Ammonia — the Energy Carrier of the Future.** Invited presentation by A. N. Simonov (online). *ICEAN 2022*, Newcastle, **Oct 2022**.
- **Electrocatalytic technologies for the sustainable synthesis of ammonia from renewables.** H.-L. Du, R. Hodgetts, D. R. MacFarlane, A. N. Simonov. *Solar Fuels Gordon Research Conference*, Tuscany, Italy, **May 2022**.
- **Engineering Scalable Electrocatalysts for Affordable Production of Green Hydrogen & E-Fuels.** Keynote presentation by A. Tricoli. *Applied Energy Symposium: MIT A+B*, Cambridge, UK, **Jul 2022**.
- **Engineering Sustainable Electrocatalysts for Renewable Energy Storage in E-Fuels.** Keynote presentation by A. Tricoli. *International Conference on Emerging Advanced Nanomaterials*, Newcastle, **Oct 2022**.
- **Towards sustainable electrosynthesis of hydrogen and ammonia.** Keynote talk by A. N. Simonov (online). *EF4 – Energy Future Conference 2021 (virtual)*, UNSW, Sydney, **Oct 2021**.
- **Towards robust, high-performance Li-mediated electrosynthesis of ammonia.** Keynote talk by A. N. Simonov (online). *240th ECS Meeting*, Orlando, USA, **Oct 2021**.
- **3D Nanostructuring of Metal-Organic Frameworks for Energy Applications.** Keynote presentation by A. Tricoli. *International Conference on Nanoscience and Nanotechnology (ICONN)*, Brisbane, **Feb 2020**.
- **Towards genuine electrocatalytic synthesis of ammonia from dinitrogen**. Invited presentation by A. N. Simonov. *Online nanoGe Fall Meeting 20 (OnlineNFM20)*, **Oct 2020**.
- **Three-dimensional Nanostructuring of Metal-Organic Frameworks for energy applications**. Keynote lecture by A. Tricoli. *ICONN - International Conference on Nanoscience and Nanotechnology*, Brisbane, **Feb 2020**.
- **Unlocking the Potential of Earth-Abundant Metal Oxides for Thermochemical Solar Fuel Production**. Invited presentation by A. Tricoli. *ACMM 26*, Canberra, **Feb 2020**.
- **Scalable Structuring of Efficient Earth-Abundant Electrocatalysts for Water Splitting and CO_2 Reduction**. Invited presentation by A. Tricoli. *ACES Symposium*, Canberra, **Feb 2020**.

- **Self-healing electrocatalysts for stable oxidation of hot acidic water**. M. Chatti, J. L. Gardiner, M. Fournier, D. R. MacFarlane, R. K. Hocking, A. N. Simonov, *Cell Symposia - Next Generation Materials for Energy Applications*, Xiamen, China **Nov 2019**.

Several presentations including an invited talk at the 237th Electrochemical Society Meeting were cancelled due to the global travel ban.

(5) Invited seminars

The CIs have delivered a range of in-person and online presentations reporting on the outcomes of the ARENA-funded project:

- in Australia: ANU, University of Western Australia, Flinders University, University of Melbourne, Royal Melbourne Institute of Technology (RMIT), University of Newcastle, Curtin University, University of New South Wales (UNSW), Monash Malaysia.
- overseas: ETH Zurich, MIT, University of Milano, GeorgiaTech, University of Bayreuth, Strasbourg University, Helmholtz Zentrum Berlin (HZB), Frei University, Fritz Haber Institute Berlin, University of Bath, Indian Institute of Technology Bombay (IITB), National University of Sciences and Technology (Pakistan), Berlin University of Technology (TU Berlin), UC Irvine, RWTH Aachen, Max Plank Institute for Chemical Energy Conversion (MPI-CEC).

Our interactions with a very broad audience of very different backgrounds through the activities above has clearly contributed to the increased awareness around the hydrogen economy and our developments in the sustainable hydrogen generation technologies among industry and general public. It was important to learn that the interest in H_2 production through electrolysis by industry has progressed to a more substantial level with a clear intention to deeply investigate future opportunities. In particular, this resulted in several consulting projects on the hydrogen production opportunities we have been involved with Australian companies.

(5) High profile, policy-level publications

These publications highlight our vision on the development of the renewable fuels technologies:

- **A Roadmap to the Ammonia Economy** by D. R. MacFarlane, P. V. Cherepanov, J. Choi, B. H. R. Suryanto, R. Y. Hodgetts, J. M. Bakker, F. M. F. Vallana, A. N. Simonov. *Joule* 2020, https://doi.org/10.1016/j.joule.2020.04.004
- **Liquefied Sunshine: Transforming Renewables into Fertilizers and Energy Carriers with Electromaterials** by D. R. MacFarlane, Choi B. H. R. Suryanto, R. Jalili, M. Chatti, L. M. Azofra, A. N. Simonov. *Advanced Materials* 2020, https://doi.org/10.1002/adma.201904804

(6) Scientific publications

1 **Mixed Silver-Bismuth Oxides: a Robust Oxygen Evolution Catalyst Operating at Low pH and Elevated Temperature.** D. Simondson, M. Chatti, J. L. Gardiner, B. V. Kerr, D. A. Hoogeveen, P. V. Cherepanov, I. C. Kuschnerus, T. D. Nguyen, B. Johannessen, S. L. Y. Chang, D. R. MacFarlane, R. K. Hocking, A. N. Simonov. *ACS Catalysis*. **2022**, *12*, 12912. https://doi.org/10.1021/acscatal.2c03065.

2 **Nanoscale TiO_2 Coatings Improve the Stability of an Earth-Abundant Cobalt Oxide Catalyst during Acidic Water Oxidation.** T. Tran-Phu, H. Chen, R. Daiyan, M. Chatti, B. Liu, R. Amal, Y. Liu, D. R. Macfarlane, A. N. Simonov, A. Tricoli. *ACS Applied Materials & Interfaces* **2022**, *14*, 33130. https://doi.org/10.1021/acsami.2c05849.

3 **Alternatives to water photooxidation for photoelectrochemical solar energy conversion and green H_2 production.** X. M. C. Ta, R. Daiyan, T. K. A. Nguyen, R. Amal, T. Tran-Phu, A. Tricoli. *Advanced Energy Materials* **2022**, 2201358. https://doi.org/10.1002/aenm.202201358.

4 **Understanding the activity and stability of flame-made Co_3O_4 spinels: A route towards the scalable production of highly performing OER electrocatalysts.** T. Tran-Phu, R. Daiyan, J. Leverett, Z. Fusco, A. Tadich, I.D. Bernardo, A. Kiy, T.N. Truong, Q. Zhang, H. Chen, P. Kluth, R. Amal, A. Tricoli. *Chemical Engineering Journal* **2022**, 429, 132180. https://doi.org/10.1016/j.cej.2021.132180.

5 **Intrinsic Catalytic Activity for the Alkaline Hydrogen Evolution of Layer-Expanded MoS_2 Functionalized with Nanoscale Ni and Co Sulfides.** E. Charnetskaya, M. Chatti, B. V. Kerr, T. Tran-Phu, T. D. Nguyen, P. V. Cherepanov, D. A. Hoogeveen, B. Johannessen, A. Tricoli, D. R. MacFarlane, R. K. Hocking, A. N. Simonov. *ACS Sustainable Chemistry & Engineering* **2022**, *10*, 7117. https://doi.org/10.1021/acssuschemeng.2c01243.

6 **Durable Electrooxidation of Acidic Water Catalysed by a Cobalt-Bismuth-based Oxide Composite: An Unexpected Role of the F-doped SnO_2 Substrate.** H.-L. Du, M. Chatti, B. Kerr, C. K. Nguyen, T. Tran-Phu, D. A. Hoogeveen, P. V. Cherepanov, A. S. R. Chesman, B. Johannessen, A. Tricoli, R. K. Hocking, D. R. MacFarlane, A. N. Simonov. *ChemCatChem* **2022**, *14*, e202200013. https://doi.org/10.1002/cctc.202200013.

7 **Nanoscale TiO_2 coatings improve the stability of an earth-abundant cobalt oxide catalyst during acidic water oxidation.** T. Tran-Phu, H. Chen, R. Daiyan, M. Chatti, B. Liu, R. Amal, Y. Liu, D. R. Macfarlane, A. N. Simonov, A. Tricoli. *ACS Applied Materials and Interfaces* **2022**, 14, 3130. https://doi.org/10.1021/acsami.2c05849.

8 **Mixed metal-antimony oxide nanocomposites: low pH water oxidation electrocatalysts with outstanding durability at ambient and elevated temperatures.** S. Luke, M. Chatti, A. Yadav, B. V. Kerr, J. Kangsabanik, T. Williams, P. V. Cherepanov, B. Johannessen, A. Tanksale, D. R. MacFarlane, R. K. Hocking, A. Alam, A. Yella, A. N. Simonov. *Journal of Materials Chemistry A* **2021**, *9*, 27468. https://doi.org/10.1039/d1ta07293e.

9 **Stable acidic water oxidation with a cobalt-iron-lead oxide catalyst operating *via* a cobalt-selective self-healing mechanism.** D. Simondson, M. Chatti, S. A. Bonke, M. F. Tesch, R. Golnak, J. Xiao, D. A. Hoogeveen, P. V. Cherepanov, J. L. Gardiner, A. Tricoli, D. R. MacFarlane, A. N. Simonov. *Angewandte Chemie International Edition* **2021**, *60*, 15821. https://doi.org/10.1002/anie.202104123.

10 **Enhancement of the photoelectrochemical water splitting by perovskite $BiFeO_3$ via interfacial engineering.** G. Liu, S. K. Karuturi, H. Chen, D. Wang, J. W. Ager, A. N. Simonov, A. Tricoli. *Solar Energy* **2020**, *202*, 198. https://doi.org/10.1016/j.solener.2020.03.117.

11 **Hybrid Organic–Inorganic Materials and Composites for Photoelectrochemical Water Splitting**. S. Singh, H. Chen, S. Shahrokhi, L. P. Wang, C.-H. Lin, L. Hu, X. Guan, A. Tricoli, Z. Xu, T. Wu. *ACS Energy Letters* **2020,** *5*, 1487. https://doi.org/10.1021/acsenergylett.0c00327.

12 **Timescale and Electrode Effects Critical for the *in situ* X-ray Spectroscopic Analysis of Electrocatalysts: the Water Oxidation Case.** H. King, M. Fournier, S. A. Bonke, E. Seeman, M. Chatti, A. N. Jumabekov, B. Johannessen, P. Kappen, A. N. Simonov, R. K. Hocking. *Journal of Physical Chemistry C* **2019**, *123*, 28533. https://doi.org/10.1021/acs.jpcc.9b06944.

13 **High Temperature One-Step Synthesis of Efficient Nanostructured $BiVO_4$ Photoanodes for Water Oxidation.** T. Tran-Phu, H. Chen, R. Bo, I. Di Bernardo, Z. Fusco, A. N. Simonov, A. Tricoli. *Energy Technology* **2019**, *7*, 1801052. https://doi.org/10.1002/ente.201801052.

14 **Intrinsically stable *in situ* generated electrocatalyst for long-term oxidation of acidic water at up to 80 °C.** M. Chatti, J. L. Gardiner, M. Fournier, B. Johannessen, T. Williams, T. R. Gengenbach, N. Pai, C. Nguyen, D. R. MacFarlane, R. K. Hocking, A. N. Simonov. *Nature Catalysis* **2019**, *2*, 457. https://doi.org/10.1038/s41929-019-0277-8.

15 **Earth-abundant transition metal oxides with extraordinary reversible oxygen exchange capacity for efficient thermochemical synthesis of solar fuels.** X. Gao, G. Liu, Y. Zhu, P. Kreider, A. Bayon, T. Gengenbach, T. Lu, Y. Liu, J. Hinkley, W. Lipiński, A. Tricoli. *Nano Energy* **2018,** *50*, 347. https://doi.org/10.1016/j.nanoen.2018.05.045.

LESSONS LEARNT AND NEXT STEPS

Our knowledge sharing activities are attracting continued attention from potential Australian and overseas partners interested in hydrogen production through water electrolysis and, in particular, in our developments in the field. From regular interactions with a range of companies we understand that the demonstration of new catalysts and electrode assemblies in a whole device is critical, which has motivated us to intensify activities towards this end. We have also gained an impression that large Australian industry are at an exploration, techno-economic analysis stage and that ongoing engagement with such analyses is important to underpin directions in electrolyser technologies.

In respect of next steps, we are enthusiastic about follow-on projects building upon the results of the current ARENA-funded work. To take the technology from its current stage of TRL 5 to TRL 8-9 and CRL 1, scale up to larger device sizes is needed to support the construction of further scalable processing and electrolysers. This would then support further detailed techno-economic analysis of the costs and benefits within the technology; this analysis would include the impact of the progressively decreasing price targets for renewables in various contexts. The latter places increasing emphasis on capital cost factors, including component lifetime; these factors are precisely those that are the focus of the technology developments in this ARENA project.

We are involved in collaborative research with Energys Australia P/L aiming to create efficient Australian water electrolysis technology. We have commenced discussions with Ixom Ltd, an Australian operator of chlor-alkali electrolysis as well as the international company ENI and Australian Government Department of Defence on possible collaborations for the creation of the hydrogen production systems for energy storage and for military needs, respectively. The latter aspect of our work has also attracted interest from the U.S. Office of Naval Research. We are at the advanced stages of planning an R&D project with the eWater systems – an Australian company developing a new hygiene system operating through water electrolysis. Finally, we have secured funding from Woodside Ltd for an R&D project (2021-2024) aimed at the development of sea water electrolysis utilising the new technologies developed herein. Our successful completion of the present ARENA-funded project has ensured that these future collaborations will significantly contribute to the development of the hydrogen technologies in Australia.

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