



**RMIT University Melbourne Hydrogen Storage and
Transport R & D Project:**

**A proton flow reactor system for electrical energy
storage and bulk export of hydrogenated carbon-
based material**

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End of Activity Report

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Acronyms and abbreviations

AC	Activated Carbon
ACPR	Activated Carbon from Phenolic Resin
ANSYS	Trade name of a computer simulation software package
C	Carbon
FTIR	Fourier-Transform InfraRed spectroscopy
GDL	Gas Diffusion Layer
H	Hydrogen
I2CNER	International Institute for Carbon Neutral Energy Research
IP	Intellectual Property
LCOE	Levelised Cost of Electricity
LOHC	Liquid Organic Hydrogen Carriers
M	Molar
MATLAB	Trade name of a mathematical solution and simulation software
MPFR	Microfluidic Proton flow reactor
O	Oxygen
PB	Proton Battery
PFR	Proton Flow Reactor
PR	Phenolic Resin
RTEE	Round Trip Energy Efficiency
sp ² , sp ³	Forms of hybridisation of electronic orbitals, by combining s and p orbitals in various linear combinations
TRL	Technology Readiness Level
V	Volt
wt	Weight
XPS	X-ray Photoelectron Spectroscopy
XRD	X-Ray Diffraction analysis

Executive Summary

This report is the End of Activity Report for the RMIT University Melbourne Hydrogen Storage and Transport R & D Project, entitled “A proton flow reactor system for electrical energy storage and bulk export of hydrogenated carbon-based material”. The aim has been to develop an integrated system for storage of electricity from renewable energy and export the stored energy as hydrogen within hydrogenated carbon-based material.

A novel ‘proton flow reactor’ (PFR) system for producing hydrogenated carbon(C)-based powder for bulk export has been developed. This reactor – a novel and scaled-up extension of RMIT’s innovative proton battery concept – uses electricity from renewables to split water and charge a stream of C-particles in a slurry electrode with the protons produced. The system is zero-emission and environmentally-benign. It offers a way to export a hydrogen-rich solid carbon material charged using renewable energy, as well as to store intermittent renewable energy on electricity grids at various scales.

It has been shown in the project that the PFR is technically feasible. There has been an experimental demonstration of the technical feasibility of all the main components of the system, including the proton flow reactor itself operating in charge and discharge modes, and the storage of hydrogen over time in dry C-particles with the liquid electrolyte used to charge them with hydrogen removed.

We have obtained for the first time direct experimental evidence that hydrogen is stored both by bonding directly to carbon atoms as well as to oxygen functional groups on carbon surfaces. Quantum-based molecular modelling work has complemented the experimental analysis by improving the understanding of the proton storage mechanisms.

The hydrogen storage capacity obtained in the so-far preferred form of activated carbon (ACPR) has been nearly tripled compared to previous results – to well over 2 wt%H. This gain is critical for the future competitiveness of the PFR system.

The potential range of applications of the system has been extended to the transportation sector by discovering a way to produce high-purity hydrogen gas directly from the hydrogenated carbon material.

Further research, development and demonstration on all the key components of the PFR system are still needed in order to improve performance and achieve the major scaling up of the system from its current small experimental scale to the very large scale required for commercial applications.

It is recommended that the experimental and molecular modelling findings on hydrogen-storage mechanisms in carbons from the present study should guide future work on synthesising carbon-based materials with properties tailored for this specific application – including activated carbons, multi-layer graphene, and layered graphitic carbon nitrides.

In a future modelling scenario in which major targeted improvements in technical performance and costs of materials and components are achieved, a PFR system for large-scale energy storage and bulk export of a hydrogen fuel could be competitive in terms of Round Trip Energy Efficiency and Levelised Cost of Electricity against both an ammonia-based system, and Liquid Organic Hydrogen Carriers.

Hence it is recommended that these assumed improvements in the simulation model become the quantitative targets for future research, development, demonstration and commercialisation activities.

A commercialisation plan to take this technology forward has been developed, with the initial focus on technology development and prototyping.

1 Introduction

This report is the End of Activity Report for the RMIT University Melbourne Hydrogen Storage and Transport R & D Project, entitled “A proton flow reactor system for electrical energy storage and bulk export of hydrogenated carbon-based material”, covering:

- Project summary and scope (section 2)
- Achievement of planned outcomes (section 3)
- Selected key highlights, and difficulties encountered (section 4)
- A summary of the current commercial status of proton flow reactor system technology (section 5), followed by a commentary on commercialisation prospects (section 6)
- Knowledge-sharing activities completed (section 7)
- Conclusions and next steps (section 8).

It is intended that this report be open for public distribution. Hence it contains no commercially-confidential information.

2 Project Summary and Scope

The aim of this project has been to develop an integrated system for storage of electricity from renewable energy and export the stored energy as hydrogen within hydrogenated carbon-based material. The project commenced in August 2018 and was completed on 19 November 2021.

A novel ‘proton flow reactor’ system for producing hydrogenated carbon(C)-based powder for bulk export has been developed. This reactor – a novel and scaled-up extension of RMIT’s innovative proton battery concept – uses electricity from renewables to split water into protons (hydronium ions) and oxygen. The protons are then stored in negatively-charged porous C-particles within a slurry electrode as it passes through the reactor.

This system offers a zero-emission and environmentally-benign solution for the production of exportable hydrogenated material from renewable energy. It may also store intermittent renewable energy on electricity grids at various scales.

RMIT’s industry partner in this project has been Eldor Corporation, Italy. The International Institute for Carbon Neutral Energy Research and Kyushu University in Japan have been academic research partners.

3 Outcomes

Outcome 1: Experimental demonstration of the practical potential of the proton flow reactor system through a technical proof-of-concept encompassing:

- **selection of C-based material producible from an abundant carbon feedstock**
- **a proton flow reactor working in charge (electrolysis) mode to store hydrogen in atomic form in this C-based powder**
- **evidence of potential capability of bulk storage and transport by tanker**
- **re-generation of electricity from the hydrogenated C-powder using the same proton flow reactor operating in discharge (fuel-cell) mode.**

A technical proof-of-concept of the proton flow reactor (PFR) system, including experimental demonstration of the feasibility of the main components of the proton flow reactor system, was achieved. A PFR was demonstrated experimentally in operation in charge and discharge modes, and the storage of hydrogen over time in dried C-particles was shown to be feasible.

The principal evidence provided that the PFR system concept is technically feasible is threefold:

- The system was experimentally demonstrated in a proton reactor with a paste electrode made by mixing activated carbon from phenolic resin (ACPR) particles in a dilute (1 molar) sulphuric acid electrolyte. A paste is a high-concentration form of slurry electrode with a near maximum density of suspended particles. After drying the charged paste electrode with argon to a near powder state, storing the powder for nearly a day, and then reforming a paste by adding liquid acid, the proton reactor used the hydrogen stored to produce electricity, that is, by working like a fuel cell. Around 60% of the hydrogen stored in the electrode after charging was recovered and consumed in the electricity generation.
- Molecular modelling using Density Functional Theory and *ab initio* molecular dynamics (AIMD) showed that reactions between hydronium (H_3O^+) and negatively-charged graphene surfaces, both with surface oxygen groups and without, can lead to hydrogen bonded to the surface, and hence effective storage of hydrogen. (AIMD is based on quantum theory, and simulates the process of chemical reactions over time.)
- Cyclic voltammograms of ACPR in acid electrolyte showed clear signs of C...H bonding. The symbol C...H is used here rather than the usual C-H, or CH, to encompass a number of possible forms of bonding including direct C-H, and C-O-H. For the PFR concept to be technically feasible, some form of bonding between H and the C atoms in the carbon material is necessary, to ensure stable storage of hydrogen after removal of the liquid acid, and over an extended period in the dry state.

Through the experimental work conducted in this project, we have extended the proof of feasibility of all the key steps in the PFR system, by showing that hydrogen can be stored in a PFR with a flow-in flow-out paste electrode, including the drying of the paste to a powder for storage externally, and reconstitution of the paste at a later time for recovery of the stored energy (Figure 1). There is a good prospect of substantially increasing the recovery rate for stored hydrogen in the carbon materials. However, further work is required to demonstrate this higher recovery rate experimentally for the case where the paste is moved to another location for drying, and in turn stored as a powder outside the PFR for a period.

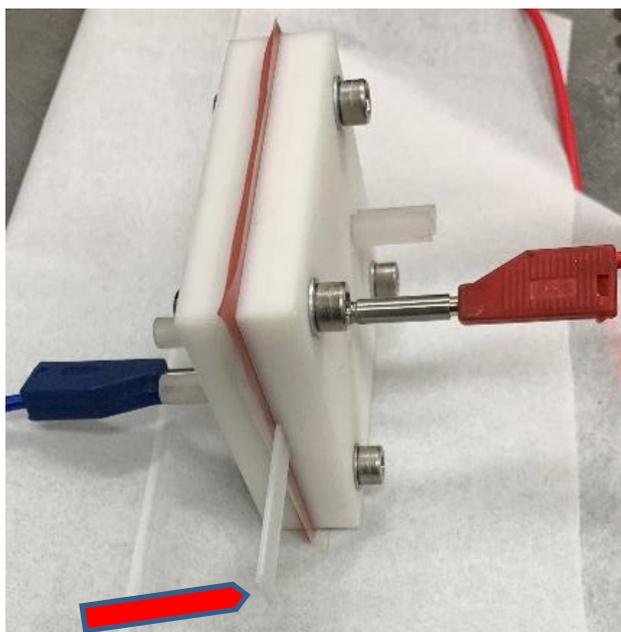


Figure 1: One of the experimental proton flow reactors made and tested. The slider used to introduce the carbon paste electrode is shown by the red arrow. The dimensions of the cell are approximately 5 x 5 x 2 cm³

A microfluidic PFR (MPFR) system has also been designed to allow direct observation of a slurry electrode through transparent sections, in situ measurement of some key performance parameters, and use of small quantities of key materials such as carbon in the slurry (Figure 2). This system was also used to test an actual carbon-based slurry electrode rather than a carbon paste electrode, and a reversible hydrogen storage of around 0.2 wt% has been obtained with an ACPR-in-acid slurry. (A storage of X wt% means that the mass of hydrogen stored is X % of the mass of the carbon material in which it is stored. While the values for X obtained are usually quite low, the energy content of the hydrogen per unit mass is high.)

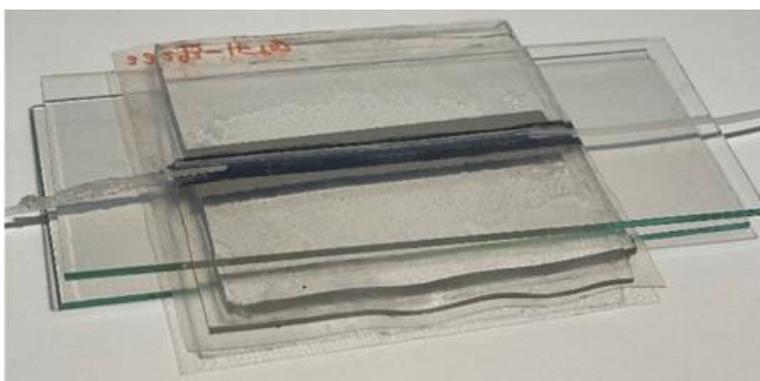


Figure 2: A microfluidic proton flow reactor with transparent ITO electrodes (dimensions 100 x 40 x 1.1 mm³)

To date we have obtained the best results for H-storage using activated carbon from phenolic resin (ACPR), but other candidate carbon-based materials have also been investigated such as Norit (the trade name of another activated carbon), selected forms of multilayer graphene, and nitrogen-doped carbons.

ACPR is currently prepared from phenol and formaldehyde, which can be made from petroleum, but also organic sources. Provided a high recycling/reuse rate is achieved for the C-based material used in

a PFR system, it should not place unsustainable demands on the primary materials required, whether petroleum, coal, or organic biomass sources.

Once the charged C-material is in powder form it can be stored and transported in bulk using well-established procedures and equipment. The C-powder would be stored in an inert gas atmosphere, such as nitrogen, and at near ambient pressure and temperature. It is not a hazardous or flammable material. All these factors will facilitate safe and economical storage and transport.

Outcome 2: Generation of performance data from an experimental scale-model of the system

The following key experimental measurements pertaining to performance of a PFR system and its components have been made:

- Experimental measurements of the performance of both a flow-in flow-out paste proton flow reactor system, and a microfluidic proton flow reactor system, in both charge and discharge modes.
- Gravimetric and volumetric energy densities (hydrogen storage capacity) of the carbon-based storage material have been measured, principally for ACPR, but also for a number of other candidate carbon-based materials such as Norit (an activated carbon), selected forms of multilayer graphene, and nitrogen-doped carbons.
- A nearly three-fold increase in the gravimetric hydrogen storage capacity in a paste-electrode proton reactor/battery employing activated carbon from phenolic resin has been achieved by heating the cell to around 70°C and using a thinner Gas Diffusion Layer (GDL) on the oxygen side. In the present project, a storage of 2.23 wt%H has been measured, compared to the best achieved before of 0.8 wt%H.
- An alternative mode of operating a PFR in discharge has been discovered that allows direct generation of hydrogen gas from the hydrogenated carbon material, thus opening up the prospect of the PFR system being used to export and supply hydrogen gas for fuel-cell vehicles for transportation.
- The energy efficiencies of the small-scale paste proton flow reactor during the hydrogen-charging (electricity supply) process, and discharging (electricity delivery) process, have been measured, and a value of 15% for the roundtrip energy efficiency for the PFR system overall obtained. Projected improvements could see that rise to 57% for a full-scale system after commercialisation.
- Some initial measurements have been made of reversible hydrogen storage of the ACPR material after multiple cycling, and after leaving in a dried charged state for up to a day. Further testing is needed to determine any storage capacity changes after multiple cycling, and with longer durations of storage within a particular cycle.
- Further work is needed to identify the best procedure to obtain a high recovery rate for the liquid electrolyte (dilute sulphuric acid) used in a paste electrode or a slurry electrode over a charge-discharge cycle.
- A thorough characterisation of the ACPR carbon electrode material using FTIR, XRD, Raman Spectroscopy, and XPS before and after charging in a proton reactor has provided direct experimental evidence for the first time that protons from hydronium are neutralised by bonding to the carbon matrix both via oxygen functional groups, and directly to carbon atoms.
- The mechanisms of formation of these C-O-H and C-H bonds observed experimentally have been elucidated by quantum-based molecular modelling studies – density functional theory and Born-Oppenheimer Molecular Dynamics – on a hydronium-water-graphene flake (charged) system.
- The main candidates for the liquid electrolyte for making the carbon paste or slurry in a PFR have been briefly reviewed, although this requires additional study in the future.

- The design of the slurry electrode in a PFR has been studied by modelling and experiments at both a microfluidic scale, and a macro scale, with the efficacy of a carbon foam current collector tested in the latter configuration.
- Technical options for drying the carbon slurry/paste after charging investigated experimentally include: low-vacuum filtration and air drying; high-vacuum filtration and fixed-bed air drying. A full setup for the latter option has been designed.
- A preliminary review of the systems available for charged carbon-particle storage and transport has been conducted.
- A demonstration model of a PFR system has been constructed to illustrate how the system works as a whole, the functions of its individual components, and the interconnections between these components. The states of the carbon medium – powder, paste, slurry – at the various stages in the overall system are shown by actual samples in transparent containers.

Outcome 3: An assessment of the energy efficiency and economic viability of a full-scale system, based on the performance of the experimental system, and a computer simulation of a full-scale system with projected impacts of economies of scale.

A simulation model of the overall proton flow reactor system for large-scale electrical energy storage and bulk export of hydrogenated carbon-based material has been constructed in MATLAB and includes representations of all the main components of the system. This model was used to estimate the round-trip energy efficiency (RTEE) of the system, that is, the electrical energy delivered after storage and transport as a percentage of the total electrical energy input, from the primary source of power (such as solar or wind power) plus process energy; and the levelised cost of electricity (LCOE) delivered after transport and storage. (LCOE is the sum of the annualised capital costs plus annual operating costs of all components and operations over a given assessment period at a selected real discount rate, divided by the annual electricity delivered.)

Two scenarios were simulated using the model:

- A base case for a small-scale PFR system with input parameters based on the best performance from our recent experiments, values obtained from the literature, and reasonable assumptions where accurate values were not obtainable at this time.
- An improved case for a future large-scale PFR with input parameters reflecting improved technical performance through technological development, cost engineering applied to all components, and economies of scale.

In the base case, the model gave a RTEE of just under 15%. This is a relatively low value, although it does partly reflect that some of the energy has potentially been stored for a long duration of up to six months. For example, some solar energy may need to be stored from mid-summer when solar radiation is at a maximum, to mid-winter when it is at a minimum, and thus the need to draw on storage is greatest. Not unexpectedly, the LCOE for the base case also comes out very high at \$22/kWh. By far the largest contributor to the LCOE was the cost of the carbon powder, with \$195/kg being assumed based on the current price of Norit activated charcoal.

In the improved case, much more efficient operation of the PFRs in both fuel cell and electrolyser modes was assumed, and generally lower losses throughout the system. The RTEE from the simulation rose to 57%, a substantially higher value. For the LCOE calculation in the improved case, the cost of carbon powder was reduced to \$1/kg, around the current price of commercial carbon black, and the costs of other components were reduced through projected technology development and economies of scale. On these assumptions, the LCOE came down to \$0.46/kWh. The major cost reductions assumed in the improved case reflect the fact that PFR technology and the required carbon materials are still at a very

early stage of technology development, and currently produced at a very small scale. While carbon black cannot itself be used as the C-material for H storage since it is not porous, it is a refined carbon-based material that is now produced in large quantities, and its production cost has fallen substantially over past years.

The RTEE and LCOE of the PFR system in the improved case would thus be competitive against both an ammonia-based system, and Liquid Organic Hydrogen Carriers (LOHCs). But realisation of the assumptions in the improved case will require major improvements both in technical performance and in costs of materials and components through technology development and economies of scale.

Outcome 4: A commercialisation roadmap developed, taking into account all the experience gained during the project

A commercialisation roadmap has been developed, in consultation with RMIT IP Commercialisation section and an industry partner

4 Key Highlights and Difficulties

Two very significant discoveries were made recently in this project that could significantly enhance the competitiveness of a PFR system.

Firstly, a nearly three-fold increase in the gravimetric hydrogen storage capacity in a paste-electrode proton reactor/battery has been achieved by making a number of modifications to the PFR design. The previously best achieved capacity for hydrogen storage in ACPR was raised from 0.8 wt% to 2.23 wt%, which is more than twice the maximum electrochemical hydrogen storages reported to date in carbons with acidic electrolytes. Specifically, the proton battery was mildly heated during discharge to a temperature of about 70°C, and the titanium gas diffusion layer was made much thinner, to enhance the water formation reaction on the oxygen-side. Further hydrogen gas evolution during charging was suppressed by limiting direct contact of hydrogen-side metals with Nafion.

Secondly, an alternative mode of operating a PFR in discharge mode has been discovered that allows direct generation of hydrogen gas from the hydrogenated carbon material. Importantly, this discovery opens up the prospect of the PFR system being used to export and supply hydrogen gas for fuel-cell vehicles for transportation, rather than solely for electrical energy storage.

Significant advances have also been made in the fundamental understanding of the reactions between protons (from hydronium H_3O^+) and negatively charged carbon surfaces that are crucial in electrochemical hydrogen storage. We have obtained for the first time direct experimental evidence that hydrogen is stored both by bonding to oxygen functional groups on carbon surfaces, as well as directly to carbon atoms, using a combination of Fourier transform infrared spectroscopy, Raman spectroscopy and X-ray photoelectron spectroscopy.

Moreover, quantum-based molecular modelling work using Density Functional Theory has complemented the experimental analysis by improving understanding of how protons are stored on oxygen-functionalised and pristine graphene flakes with excess negative charge.

A firm basis for eventual commercialisation of the PFR system and its components has been laid by filing on 25 June 2021 a Patent Cooperation Treaty application covering the entire PFR system, and

the design enhancements recently achieved. This application followed the provisional patent (Australia) for the system that was obtained in June 2020.

By far the most serious difficulty we have experienced in the conduct of this project has been the limitation on access to RMIT University laboratories due to the extensive Victorian Government covid-19 lockdowns that have been in place during 2020 and 2021. These lockdowns have inevitably reduced the amount of experimental work that has been able to be done. Many design improvements identified in the PFR system and its components, and in the basic carbon hydrogen storage material, were thus not able to be pursued as far as originally expected within the present project. But we have in place well-developed plans to realise these improvements in future research, development and demonstration on this system.

5 Commercial Status of Project Technology

This project has achieved its goal of taking proton flow reactor technology from Technology Readiness Level (TRL) 2 to 4, so that the system and its main components have now been demonstrated at a small scale in experimental systems. There is still a long way to progress from the small-scale experimental systems that have been built and tested, to large-scale commercial systems, particularly a system that could store hydrogen in a carbon-based material in bulk and then export this in tankers to overseas locations.

The central technological challenge facing PFR system technology is thus to prove that this scale-up is both technically and economically feasible.

6 Commentary on Commercialisation Prospects

In the system modelling done as part of this project, estimates have been made of the round-trip energy efficiency (RTEE) of the system, and the levelised cost of electricity (LCOE) delivered after transport and storage. A scenario of a future large-scale PFR was examined with input parameters reflecting improved technical performance through technological development, cost engineering applied to all components, and economies of scale.

Importantly, in this scenario, the RTEE rose to 57%, and the LCOE came down to \$0.46/kWh. At these values, the PFR system would be competitive against both an ammonia-based system, and Liquid Organic Hydrogen Carriers (LOHCs). But realisation of the assumptions in this scenario will require major improvements both in technical performance and in costs of materials and components through the associated technology development and economies of scale.

These assumed levels of technical performance and unit costs in this scenario thus provide clear targets for future research, development, demonstration and prototyping activities that need to be met to achieve competitiveness for the PFR system, and hence to realise the prospects for full commercialisation.

The potential range of applications, and hence markets, for PFR system technology if it can become competitive are vast and include:

- From household to community, commercial and industrial scale electrical energy storage for solar photovoltaic systems

- Grid-scale electrical energy storage to allow integration of major proportions of intermittent solar and wind power
- Export of hydrogen in the form of the hydrogenated carbon solid-state fuel, charged using Australia's abundant renewable energy resources
- Use of this fuel in importing countries for electricity generation, or for supply of high-purity hydrogen gas to fuel-cell vehicles in the transport sector,

All of these applications are zero greenhouse gas emission routes, and this support the global effort to achieve net zero emissions by 2050.

A realistic quantitative assessment of market prospects will need to await the results of the next stages of technology development to test the performance of the PFR system at much larger scales than have so far been realised.

7 Knowledge-sharing Activities Completed

The main knowledge-sharing activities conducted since the project commenced are detailed in Table 1. The list of publications arising from the project is presented in Table 2.

Researcher	Date	Knowledge sharing activity	Event/organisation	Location
Professor John Andrews	6-9 November 2018	Technical visit and presentation (8 November 2018)	International Institute for Carbon Neutral Energy Research (I2CNER) and Kyushu University	Fukuoka, Japan
		Presentation video	Made available via YouTube by I2CNER	www.youtube.com/watch?v=UIXzhE7KS18
	12 November 2018	Technical visit and discussions	Eldor Corporation (industry partner) Research Laboratory.	Orsenigo, Italy
	16 November 2018	Presentation to President and senior management, and discussions	Eldor Corporation (industry partner), President's residence,	Castiglione d'Orcia, Italy
	20 November 2018	Technical visit	Precors (fuel cell bipolar plate manufacturer)	Julich, Germany
	22 November 2018	Technical visit and presentation	Department of Chemistry, University of Cambridge	Cambridge, UK
RMIT ARENA project research team	20 November 2019	Technical workshop	I2CNER and Kyushu University, Japan, visit by Professors H. Matsumoto and T.Fujigaya and PhD student	RMIT Melbourne
RMIT ARENA research team	27 November 2019	Video presentation on progress	Eldor Corporation	Melbourne/Italy
Professor Gary Rosengarten	20 June 2019	Technical visit and discussions	Prof. Jean-Luc Meunier, Plasma Processing Laboratory, Dept of Chemical Engineering, McGill University leading to sending sample carbon nanotubes being sent	Montreal, Canada

			for testing stability, conductivity and H-storage potential	
Dr Seyed Niya	4 December 2019	Conference presentation	Australian Nano and Energy Materials 2019 Conference	University of Western Australia, Perth
Prof. John Andrews and Prof. Gary Rosengarten	2019-2020	Project description	RMIT website	https://www.rmit.edu.au/research/our-research/enabling-capability-platforms/information-systems-engineering/energy/research-areas/conversion-and-storage
RMIT/Prof. John Andrews, Prof. Gary Rosengarten, Dr Shahin Heidari, Sr Seyed Niya, Saeed Seif Mohammadi, Francois du Toit	June 2020	Provisional patent application lodged with IP Australia	"Proton Flow Reactor System"	Melbourne
Prof. John Andrews	Feb. 2021	Presenting the proton flow reactor and proton battery innovations to CSIRO hydrogen mission program leader, Dr Patrick Hartley		Online Forum, RMIT
Prof. John Andrews A/Prof. Bahman Shabani	April 2021	Presenting the proton flow reactor and proton battery innovations to over 100 highly influential members of the Australian hydrogen community (AHC, DELWP, regional hydrogen clusters, CSIRO) and Dr Alan Finkel, former chief scientist of Australia	Hydrogen house @ RMIT	RMIT City Campus and online https://www.rmit.edu.au/events/2021/april/hydrogen-house-at-rmit
Prof. John Andrews	May 2021	Presentation on "URFC, proton battery, and proton flow reactor energy storage systems"	Australian Hydrogen Research Network	Online seminar
RMIT ARENA Project Research Team	June 2021	Patent Cooperation Treaty (PCT) application on 'Proton Flow Reactor System' filed on 25 June 2021 (PCT/AU2021/050670), following the provisional patent application one year earlier	Australian Patent Office	Electronic submission
A/Prof. Bahman Shabani	Sep. 2021	Presenting the proton flow reactor and proton battery innovations to ~400 members of Engineers Australia	Organised by Advisian and Engineers Australia	Online Event https://www.engineersaustralia.org.au/event/2021/07/green-hydrogen-energy-storage-and-transport-38086
Professor John Andrews	18-20 Oct 2021	Invited Presentation	4 th Energy Future Conference	Fully Virtual https://www.ausenergyfuture.com/

Professor Michelle Spencer	18-20 Oct 2021	Invited Presentation	4 th Energy Future Conference	Fully Virtual https://www.ausenergyfuture.com/
RMIT ARENA Project Research Team	December 2021	Workshop to present findings of this project to the wider academic, industry, public sector, and general communities	On-line workshop	Currently being planned.

Table 1: Knowledge-sharing activities conducted since the project commenced

Published Journal Papers

1. M. Mourshed, S.M.R. Niya, R. Ojha, G. Rosengarten, J. Andrews, B. Shabani (2021) *Carbon-based slurry electrodes for energy storage and power supply systems*, Energy Storage Materials (impact factor: **17.8**), 40, 461-489 (<https://doi.org/10.1016/j.ensm.2021.05.032>).
2. J. Andrews, S.M.R. Niya, R. Ojha (2022) *Electrochemical hydrogen storage in porous carbons with acidic electrolytes: Uncovering the potential*, Current Opinion in Electrochemistry (impact factor: **7.3**), 31, 100850 (<https://doi.org/10.1016/j.coelec.2021.100850>).
3. A. Heidarian, S. Cheung, R. Ojha, G. Rosengarten (2022) *Effects of current collector shape and configuration on charge percolation and electric conductivity of slurry electrodes for electrochemical systems*, Energy (impact factor: **7.1**), 239, 122313 (<https://doi.org/10.1016/j.energy.2021.122313>).
4. S.M.R. Niya, J. Andrews (2022) *On charge distribution and storage in porous conductive carbon structure*, Electrochimica Acta (impact factor: **6.9**), In-press (<https://doi.org/10.1016/j.electacta.2021.139534>).
5. J. Andrews, R. Ojha, S.M.R. Niya, S. Seibt (2022) *Electrochemical storage reactions of hydrogen in activated carbon from phenolic resin*, Catalysis Today (impact factor: **6.8**), In-press.
6. A. Heidarian, S. Cheung, G. Rosengarten (2021) *The effect of flow rate and concentration on the electrical conductivity of slurry electrodes using a coupled computational fluid dynamic and discrete element method (CFD-DEM) model*, Electrochemistry Communications (impact factor: **4.7**), 126, 107017 (<https://doi.org/10.1016/j.elecom.2021.107017>).

Peer-reviewed Conference Presentations

1. J. Andrews, R. Ojha, S.M.R. Niya (2021) *Electrochemical storage reactions of hydrogen in activated carbon from phenolic resin*, Fifth Edition of Catalysis and Chemical Engineering, February 22-24, California, USA.
2. S.M.R. Niya, S. Heidari, F. Du Toit, G. Rosengarten, J. Andrews (2019) *Electrochemical storage of hydrogen in carbons: Opportunities for bulk storage*, Advanced Nano and Energy Materials (ANEM2019), December 4-6, The University of Western Australia, Perth, Australia.

Papers In-preparation

1. S.M.R. Niya, S. Heidari, J. Andrews (2022) *Enhanced electrochemical hydrogen storage in an improved proton battery*, Soon-to-be submitted.
2. M. Mourshed, S.M.R. Niya, B. Shabani (2022) *Analysis of the electronic and ionic conductivity of a carbon-based slurry electrode: effects of slurry concentration, flow channel geometry, and flow rate*, Internal reviewing level.
3. A. Heidarian, S. Cheung, G. Rosengarten (2022) *On the effects of particle size and shape of suspended particles on viscosity and conductivity of slurry electrode*, First draft preparation.
4. J. Andrews, G. Rosengarten, S.M.R. Niya, R. Ojha, S. Heidari, F. du Toit, S. Cheung, B. Shabani, S. Spencer (2022) *Proton flow reactor system for electrical energy storage and bulk export of hydrogenated carbon-based material*, Paper design level.

Table 2: Publications arising from the project

8 Conclusions and Next Steps

The most important conclusion of this project is that a proton flow reactor system for potential use for electrical energy storage and export of hydrogen in the form of a hydrogenated carbon-based material is technically feasible. There has been an experimental demonstration of the technical feasibility of the main components of the proton flow reactor system, including the proton flow reactor itself operating in charge and discharge modes, and the storage of hydrogen over time in dry carbon particles with the liquid electrolyte used to charge them with hydrogen removed.

We have obtained for the first time direct experimental evidence that hydrogen is stored both by bonding to oxygen functional groups on carbon surfaces, as well as directly to carbon atoms. Quantum-based molecular modelling work has complemented the experimental analysis by improving the understanding of the proton storage mechanisms.

The hydrogen storage capacity obtained in the so-far preferred form of activated carbon (ACPR) has been nearly tripled compared to previous results – to well over 2 wt%H. This gain is critical for the future competitiveness of the PFR system.

The potential range of applications of the system has been extended to the transportation sector by discovering a way to produce high-purity hydrogen gas directly from the hydrogenated carbon material.

Further research, development and demonstration on all the key components of the PFR system are still needed in order to improve performance and achieve the major scaling up of the system from its current small experimental scale to the very large scale required for commercial applications.

On the fundamental question of what carbon-based material is the most suitable for this system, it is recommended that the experimental and molecular modelling findings on hydrogen-storage mechanisms in carbons from the present study should guide future work on synthesising carbon-based materials – including activated carbons, multi-layer graphene, and layered graphitic carbon nitrides – that have properties tailored for this specific application.

In a future modelling scenario in which major improvements both in technical performance and in costs of materials and components through technology development and economies of scale are achieved, a PFR system for large-scale energy storage and bulk export of a hydrogen fuel could be competitive in terms of Round Trip Energy Efficiency and Levelised Cost of Electricity against both an ammonia-based system, and Liquid Organic Hydrogen Carriers.

Hence it is recommended that these assumed improvements in the simulation model should become the quantitative targets for future research, development, demonstration and commercialisation activities.

A commercialisation plan to take this technology forward has been developed, with the initial focus on technology development and prototyping.