RGC Ref.: X_HKU710/14
(please insert ref. above)

The Research Grants Council of Hong Kong
SFC/RGC Joint Research Scheme
Completion Report

(Please attach a copy of the completion report submitted to the Scottish Funding Council by the Scottish researcher)

Part A: The Project and Investigator(s)

1. Project Title

Development of novel electrochemical cells for high-efficiency conversion of carbon dioxide to fuels

2. Investigator(s) and Academic Department/Units Involved

	Hong Kong Team	Scottish Team
Name of Principal Investigator *(with title)*	Prof. Dennis Y.C. Leung	Prof. Mercedes Maroto-Valer
Post	Professor	Professor and Head of institute
Unit / Department / Institution	Department of Mechanical Engineering, the University of Hong Kong	Institute of Mechanical, Process and Energy Engineering, Heriot Watt University
Contact Information	Email: ycleung@hku.hk	Email: M.Maroto-Valer@hw.ac.uk
Co-investigator(s) *(with title and Institution)*	Prof. Michael K.H. Leung, City University of Hong Kong	Drs. H.Z. Wang & J. Xuan, Heriot Watt University

3. Project Duration

	Original	Revised	Date of RGC/ Institution Approval *(must be quoted)*
Project Start date	1-1-2015		1-1-2015
Project Completion date	31-12-2015		31-12-2015*
Duration *(in month)*	12		12
Deadline for Submission of Completion Report	31-12-2016		31-12-2016

*Project completion date is 31st Dec. 2015 for the HK team and 30th June 2016 for the Scottish team.

Part B: The Completion Report

5. Project Objectives

5.1 Objectives as per original application

1. *To establish a long-term research collaboration between Heriot-Watt University (HWU) and the University of Hong Kong (HKU) to develop electrochemical fuel cells for high-efficiency CO2 reduction;*
2. *To synthesize electro-catalysts (i.e., Ag/graphene and Ag/TiO2/graphene) with controlled particle size and chemical composition;*
3. *To test electrochemical performance of the synthesized materials in both half-cell and full-cell setups;*

4. *To study impacts of structural parameters (e.g., porosity, particle size, catalytic layer thickness) on kinetics and transport;*
5. *To optimize the composition and structure of electrodes for high-efficiency and high-rate CO2-to-CO conversion.*

5.2 Revised Objectives

Date of approval from the RGC: _____

Reasons for the change: _____

_____

6. Research Outcome

Major findings and research outcome
(maximum 1 page; please make reference to Part C where necessary)

The SEM and TEM characterization showed that the size and density of PdNP particles loaded in the GA array, and its corresponding mass loading was gradually increased upon the increase of the initial Pd(VI) ion concentration in the raw dispersion. The GA also showed high porosity based on SEM observations. Elemental analysis from XPS even showed that the NF just act as substrate.

In the electrochemical analysis, the DMFC and DEFC half-cell scaled cyclic voltametric (CV) analysis showed that overall activity of binder-free Pd/GA/NF electrode increased when the Pd loading (weight percentage) in the electrode increased from 0.8 wt% to 7.65 wt%. Activity in ethanol oxidation was better than that in methanol from the single cycle CV operation after the catalyst activation.

Further analysis in the prolong service of the binder-free electrode was also carried out. The variation in the peak current recorded in the anodic sweep, and the corresponding toxic tolerance towards carbon monoxide showed that 7.65 wt%-Pd/GA/NF had better performance in ethanol throughout a prolong service of around 15 h.

On the device side, after conducting a systematic investigation to apply pH differential technique on a microfluidic CO_2 conversion reactor, we come to the conclusion that catholyte pH=2 and anolyte pH=14 output the optimal cell performance. In a dual electrolyte system, the reactivity was tripled than a single neutral electrolyte arrangement and the peak Faradaic efficiency was improved from 81.6% to 95.6%. Parametric study demonstrates the optimal catalyst to Nafion ratio of 30:1, electrolyte flow rate of 500 μL/min, and channel thickness of 100 μ m. Experiment on CO_2 supply rate implies no significant improvement when reaching 50 mL/min. In addition, the cell durability was assessed with regard to its repetitiveness and long period operation. After 30,000 second operation, the reactivity rate and Faradaic efficiency end up with a degradation of 41% and 11.5%, respectively.

Potential for further development of the research and the proposed course of action *(maximum half a page)*

The strong activity exhibited by the Pd/GA/NF in the half-cell reaction indicates that the applications in the full-cell stack operations, such as DFEC and glucose cells, can be further developed. The GA/NF based products can also be adapted for the indoor air quality (IAQ) application and the adsorption of waste oils from the wastewater.

The effect of operation temperature in a pH-differential microfluidic reactor is worth further studying. Researches on membrane-based reactor have indicated that the elevated reaction temperature would indeed favor the intrinsic selectivity for formate generation over hydrogen evolution, whilst adverse effects would also exist on CO_2 solubility and hence lower the intrinsic kinetics because of CO_2 mass transfer constraint. The consequence of the opposing facts was a net decrease of formate generation with higher temperature and this dilemma should be studied specifically for the microfluidics network.

7. The Layman's Summary

(describe in layman's language the nature, significance and value of the research project, in no more than 200 words)

Issues related to CO_2 have become a focus of global attention because of its emission explosion since industrial revolution and consequent adverse effect on climate. It has now become a consensus that immediate measures must be carried out to intensively reduce environmental CO_2 content. As an effective strategy of tackling the greenhouse gas issue, electrochemical utilization of CO_2 has attracted more and more attention because of its mild operation condition and high efficiency.

This project aims at gaining a thorough understanding of the electrode processes and developing general principles of electrode design for high-performance CO_2-to-CO conversion. The proposed study will focus on synthesizing porous electrode materials, characterizing their electrochemical performance for CO_2-to-CO conversion, and understanding the catalyst structure- cell performance relationship. A coupled experimental and modelling approach will be applied to achieve the insights into the transport and reactions within micro-structured electrodes. The outputs of this study will serve as the support for our second phase research in which the well-designed high-performance electrode will be integrated into an optimized reactor for CO_2 conversion at maximum performance.

Part C: Research Output

8. Peer-reviewed journal publication(s) arising directly from this research project

(Please attach a copy of each publication and/or the letter of acceptance if not yet submitted in the previous progress report(s). All listed publications must acknowledge RGC's funding support by quoting the specific grant reference.))

The Latest Status of Publications				Author(s) *(**bold** the authors belonging to the project teams and denote the corresponding author with an asterisk*)*	Title and Journal/ Book *(with the volume, pages and other necessary publishing details specified)*	Submitted to RGC *(indicate the year ending of the relevant progress report)*	Attached to this report *(Yes or No)*	Acknowledged the support of this Joint Research Scheme *(Yes or No)*	Accessible from the institutional repository *(Yes or No)*
Year of publication	Year of Acceptance *(For paper accepted but not yet published)*	Under Review	Under Preparation *(optional)*						
2016				**X. Lu, J. Xuan, DYC Leung,*** HY Zou, J. Li, H.Wang and **HZ Wang**	A switchable pH-differential unitized regenerative fuel cell with high performance. Journal of Power Sources 314, 76-84, 2016		Yes	Yes	Yes
2016				**X. Lu, DYC Leung,* HZ Wang, M. Mercedes Maroto-Valer, J. Xuan**	A pH-differential dual-Electrolyte Microfluidic Electrochemical Cells for CO_2 Utilization. Renewable Energy 95, 277–285, 2016.		Yes	Yes	Yes

9. Recognized international conference(s) in which paper(s) related to this research project was/were delivered *(Please attach a copy of each delivered paper. All listed papers must acknowledge RGC's funding support by quoting the specific grant reference.)*

Month/Year/ Place	Title	Conference Name	Submitted to RGC *(indicate the year ending of the relevant progress report)*	Attached to this report *(Yes or No)*	Acknowledged the support of this Joint Research Scheme *(Yes or No)*	Accessible from the institutional repository *(Yes or No)*

Nov/2015/ Fuzhou, China	A Switchable pH-differential Reactor with High Reactivity and Efficiency for CO_2 Utilization	CUE2015-Applied Energy Symposium and Summit 2015: Low carbon cities and urban energy systems		Yes	Yes	Yes

10. Student(s) trained *(Please attach a copy of the title page of the thesis.)*

Name	Degree registered for	Date of registration	Date of thesis submission/ graduation
Mr. X. Lu	PhD	1-9-2013	15-8-2016*

*Copy of the title page of the thesis attached.

11. Other impact *(e.g. award of patents or prizes, collaboration with other research institutions, technology transfer, etc.)*

A full proposal (Development of a pH-differential laminar flow based carbon neutral energy conversion system) based on the result of the project was submitted to RGC CERG for funding in 2015-16. Result: 3.5 (not funded)