RGC Ref.: N_PolyU528/16
NSFC Ref.: 51661165013

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

(Please attach a copy of the completion report submitted to the NSFC by the Mainland researcher)

Part A: The Project and Investigator(s)

1. Project Title

High-capacitance Wearable Li-ion Batteries using Multi-shelled Metal Oxides and Metallic Textile Electrodes

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

	Hong Kong Team	Mainland Team
Name of Principal Investigator *(with title)*	Prof. Zijian ZHENG	Dr. Mei YANG
Post	Professor	Associate Professor
Unit / Department / Institution	Institute of Textiles and Clothing / The Hong Kong Polytechnic University	Institute of Process Engineering / Chinese Academy of Sciences
Contact Information	zijian.zheng@polyu.edu.hk	myang@ipe.ac.cn
Co-investigator(s) *(with title and institution)*	N.A.	Dr. Nan XU, Dr, Jian QI & Ms Dan MAO / Institute of Process Engineering / Chinese Academy of Sciences

3. Project Duration

	Original	Revised	Date of RGC/ Institution Approval *(must be quoted)*
Project Start date	1 Jan 2017	N.A.	1 Jan 2017
Project Completion date	31 Dec 2020	N.A.	31 Dec 2020
Duration *(in month)*	48	N.A.	48
Deadline for Submission of Completion Report	31 Dec 2021	N.A.	31 Dec 2021

Part B: The Completion Report

5. Project Objectives

5.1 Objectives as per original application

1) To prepare metallic textiles with high conductivity and flexibility;
2) To synthesize and regulate metal oxide spheres with multi-shelled structure;
3) To design and fabricate flexible composite electrodes and assemble full-cell devices;
4) To fabricate and study high-capacitance Li-ion batteries using as-made composite electrodes;
5) To demonstrate the applications of textile-based Li-ion batteries in wearable electronic devices.

5.2 Revised Objectives

N.A.

6. Research Outcome

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

Metallic textile fabrics and metal oxides have been successfully prepared, which could be utilized for the assembly of textile-based composite electrodes. Textile-based Li-ion batteries have been demonstrated, exhibiting promising electrochemical and mechanical performance which are suitable for powering flexible electronics and smart garments. Detailed research outcomes are listed as follows.

1) Both Cu-coated and Ni-coated fabrics showed exceedingly low sheet resistance of 0.1 – 0.5 Ω cm^{-2} with outstanding mechanical stability after 10,000 bending cycles, being promising to serve as conductive current collectors for flexible batteries. Moreover, these metallic fabrics exhibited excellent tensile, compression strength, and washability, which could satisfy the flexible and wear-resistant requirements for wearable applications. A new reversed opal templating method was developed to fabricate highly porous metallic current collectors, which showed coating of metal on the surface of the inner pores.

2) Various kind of metal oxide microspheres with multi-shelled structures, including V_2O_5, Fe_2O_3, Co-Mn, Li-Ni-Mn-Co, TiO_2/Fe_2TiO_5, $Fe_2(MoO_4)_3$, NiO and NiS_2, were successfully synthesized. By controlling the parameters of the synthesizing processes, multi-shelled metal oxide microspheres with different compositions and shell numbers (up to 7) could be developed. Their fabrication approaches and parameters that could affect the material properties have been systematically studied.

3) Several types of composite electrodes, i.e., V_2O_5/Ni-fabric, NiO/Ni-fabric, $Ni(OH)_2$/Ni-porous substrates, Li/Cu-fabric, polyS/Ni-fabric were prepared. Commercial materials such as LFP and LTO were also used to fabricate composite electrodes for form LFP/Ni-fabric, and LTO/Cu-fabric. Such electrodes exhibited the outstanding specific capacity, rate performance and retention capability. Several ones broke the state-of-the-art performances.

4) Several kinds of flexible Li batteries including V_2O_5//Li, NiO//Li, S//Li, LFP//LTO, NiOH//C have been demonstrated. By utilizing the composite structures, these full-cells showed high areal capacity, high specific capacity, high energy density, long cycle life,. These full cells can maintain stable charge/discharge characteristics while being repeatedly bent at small radii of curvature, showing the great potential in powering flexible electronics and smart garments.

5) We advocated figure of merit of flexible batteries, as a single value metric to evaluate the performance of flexible battery. This is the first time one provides a detail discussion about how to take flexibility and energy density-two key factors that usually conflict-into consideration together, so as to truly reflect the performance of a flexible battery.

During the project period, both Hong Kong and Mainland teams organized several meetings and laboratory visits, in 2017, 2018 and 2019. These works have been published in 14 papers in high-impact journals such as Advanced Materials, Advanced Energy Materials, Joule, Nature Communications, and 2 patent filings. The results were presented in more than 10 conferences and seminars. The detail outcomes are listed in Section C below. One joint PhD student was trained.

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

Various textile-based Li-ion batteries have been developed, displaying excellent flexibility and cycling capability, which are critical in wearable applications. Their storage capacity and energy density, however, are still limited. Improvement on their electrochemical performance is required, thereby meeting the market needs prior to further development. One possible development avenue is to reduce the inert components of the cells by developing thinner and lighter building blocks of the textile current collectors and by increasing the packing density of the active materials.

Moreover, in order to put the research projects into practice, academy-industrial R&D cooperation would be established in future. Further technology transfer of project results to industry would also be feasible through the creation of spin-off.

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)

Wearable electronics, like smartwatches and health trackers, are increasingly integrated into our daily life. This ever-growing demand has driven increased interest in flexible batteries for better wearing comfort. However, flexible batteries, compared with their rigid counterparts, usually demonstrate poor performance.

In this project, high-performance flexible lithium batteries have been developed. Relying on textile-based electrodes coupled with active electrode materials and conductive metallic fabrics, the battery can store more energy and perform in a stable and durable way. Taking advantage of the desired structure of metal oxide and the unique features of textiles, the batteries can be created with high capacities and great flexibility. These batteries maintain constant power output even when folded, twisted, squeezed, and impacted, emphasizing the technology's promise for application in flexible and wearable applications.

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)*						
2021	/	/	/	**Yuqi Zhang**, **Wenshuo Wang**, **Lei Wang**, **Qianyi Guo**, **Hong Hu**, **Chuan Xie**, **Jian Shang**, **Junling Xu**, **Yaokang Zhang**, **Zijian Zheng***	Title: Inverse Opaline Metallic Membrane Addresses the Tradeoff Between Volumetric Capacitance and Areal Capacitance of Supercapacitor Journal: *Advanced Energy Materials* 2021, 2102802 (DOI: 10.1002/aenm.202102802)	2021	Yes	Yes	Yes
2021	/	/	/	**Jian Chang**, **Hong Hu**, **Jian Shang**, Ruopian Fang, Dahua Shou, **Chuan Xie**, **Yuan Gao**, **yu Yang**, **Qiuna Zhuang**, **Xi Lu**, **Yaokang Zhang**, Feng Li, **Zijian Zheng***	Title: Rational Design of Li-Wicking Hosts for Ultrafast Fabrication of Flexible and Stable Lithium Metal Anodes Journal: *Small* 2021, 2105308 (DOI: 10.1002/smll.202105308)	2021	Yes	Yes	Yes

2021	/	/	/	**Yufeng Luo**, **Yuan Gao**, **Qianyi Guo**, **Zijian Zheng***	Title: Interfacial design of thick sulfur cathodes to achieve high energy density and stability Journal: *Journal of Materials Chemistry A* 2021, 9, 17129-17142 (DOI: 10.1039/D1TA04059F)	2021	Yes	Yes	Yes
2021	/	/	/	**Dongrui Wang**, **Jian Chang**, **Qiyao Huang***, **Dongdong Chen**, **Peng Li**, Yau-Wai Denis Yu, **Zijian Zheng***	Title: Crumpled, high-power, and safe wearable Lithium-Ion Battery enabled by nanostructured metallic textiles Journal: *Fundamental Research* 2021, 1, 399-407 (DOI: 10.1016/j.fmre.2021.06.007)	2021	Yes	Yes	Yes
2021	/	/	/	**Wenshuo Wang**, **Qi Gan**, **Yuqi Zhang**, **Xi Lu**, Huixin Wang, **Yaokang Zhang**, **Hong Hu**, **Lina Chen**, Lianxin Shi, Shutao Wang, **Zijian Zheng***	Title: Polymer-Assisted Metallization of Mammalian Cells Journal: *Advanced Materials* 2021, 33, 2102348 (DOI: 10.1002/adma.202102348)	2021	Yes	Yes	Yes

2021	/	/	/	**Jian Chang**, **Qiyao Huang**, **Yuan Gao**, **Zijian Zheng***	Title: Pathways of Developing High-Energy-Density Flexible Lithium Batteries Journal: *Advanced Materials* 2021, 33, 2004419 (DOI: 10.1002/adma.202004419)	2021	Yes	Yes	Yes
2021	/	/	/	**Yuan Gao**, **Chuan Xie**, **Zijian Zheng***	Title: Textile Composite Electrodes for Flexible Batteries and Supercapacitors: Opportunities and Challenges Journal: *Advanced Energy Materials* 2021, 11, 2002838 (DOI: 10.1002/aenm.202002838)	2021	Yes	Yes	Yes
2020	/	/	/	**Jian Chang**, **Qiyao Huang**, **Zijian Zheng***	Title: A Figure of Merit for Flexible Batteries Journal: *Joule* 2020, 4, 1346-1349 (DOI: 10.1016/j.joule.2020.05.015)	2021	Yes	Yes	Yes

2020	/	/	/	Chunlei Jiang, Lei Xiang, Shijie Miao, Lei Shi, Donghao Xie, Jiaxiao Yan, **Zijian Zheng***, Xiaoming Zhang, Yongbing Tang*	Title: Flexible Interface Design for Stress Regulation of a Silicon Anode toward Highly Stable Dual-Ion Batteries Journal: *Advanced Materials* 2020, 32, 1908470 (DOI: 10.1002/adma.201908470)	2021	Yes	Yes	Yes
2020	/	/	/	Yujing Zhu, Jiangyan Wang, **Chuan Xie**, **Mei Yang**, **Zijian Zheng***, Ranbo Yu*	Title: Hollow multishelled structural NiO as a "shelter" for high-performance Li–S batteries Journal: *Materials Chemistry Frontiers* 2020, 4, 2971-2975 (DOI: 10.1039/D0QM00492H)	2021	Yes	Yes	Yes
2020	/	/	/	Yujing Zhu, **Mei Yang**, **Qiyao Huang**, **Dongrui Wang**, Ranbo Yu*, Jiangyan Wang, **Zijian Zheng***, Dan Wang*	Title: V_2O_5 Textile Cathodes with High Capacity and Stability for Flexible Lithium-Ion Batteries Journal: *Advanced Materials* 2020, 32, 1906205 (DOI: 10.1002/adma.201906205)	2021	Yes	Yes	Yes

2020	/	/	/	**Qianyi Guo**, **Zijian Zheng***	Title: Rational Design of Binders for Stable Li-S and Na-S Batteries Journal: *Advanced Functional Materials* 2020, 30, 1907931 (DOI: 10.1002/adfm.201907931)	2021	Yes	Yes	Yes
2019	/	/	/	Jing Wang, **Mei Yang***, **Zijian Zheng**, Ranbo Yu*, Dan Wang*	Title: Design, preparation and assembly of flexible electrode based on carbon materials Journal: Chinese Science Bulletin 2019, 64, 514-531 (DOI: 10.1360/N972018-01105)	2018	No	Yes	Yes
2018	/	/	/	**Jian Chang**, **Jian Shang**, Yongming Sun, Luis K. Ono, **Dongrui Wang**, **Zhijun Ma**, **Qiyao Huang**, **Dongdong Chen**, **Guoqiang Liu**, Yi Cui, Yabing Qi & **Zijian Zheng***	Title: Flexible and stable high-energy lithium-sulfur full batteries with only 100% oversized lithium Journal: Nature Communications 2018, 9, 4480 (DOI: 10.1038/s41467-018-06879-7)	2018	No	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)*
10/2018/ Qingdao	Multi-shelled Fe2O3 wrapped carbon nanotubes as a high-capacitanc e flexible electrode for lithium ion batteries	Asia NANO 2018	2018	No	Yes	No
10/2018/ Qingdao	Fiber-enabled Wearable Electrochemical Storage Devices	Asia NANO 2018	2018	No	Yes	No
05/2019/ Nice/France	Fiber-enabled Wearable Energy Storage Devices	European Materials Research Society	2021	Yes	Yes	No
12/2019/ Jeju	Fiber-enabled Wearable Energy Storage Devices	The 11th International Conference on Advanced Materials and Devices (ICAMD2019)	2021	Yes	Yes	No

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
Yujing ZHU	PhD	10/Sep/2015	16/Jun/2020

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

US provisional patents:

1) Nanostructured Metallic Textiles for Current Collectors of Flexible Lithium Ion Battery (Patent no.: 62/610,977)
2) Multifunctional Hierarchical Fabrics Stabilize Flexible Lithium Metal-Sulfur Batteries (Patent no.: 62/599,815)

12. Statistics on Research Outputs *(Please ensure the summary statistics below are consistent with the information presented in other parts of this report.)*

	Peer-reviewed journal publications	Conference papers	Scholarly books, monographs and chapters	Patents awarded	Other research outputs (Please specify)
No. of outputs arising directly from this research project [or conference]	14	4	0	2	>10 invited presentations in institutions