RGC Ref. No.:
UGC/FDS25/E06/19
(please insert ref. above)

RESEARCH GRANTS COUNCIL
COMPETITIVE RESEARCH FUNDING SCHEMES FOR
THE LOCAL SELF-FINANCING DEGREE SECTOR

FACULTY DEVELOPMENT SCHEME (FDS)

Completion Report
(for completed projects only)

Submission Deadlines:
1. *Auditor's report with unspent balance, if any: within **six** months of the approved project completion date.*
2. *Completion report: within **12** months of the approved project completion date.*

Part A: The Project and Investigator(s)

1. Project Title

Development of composite separators with superior thermal characteristics for safety enhancement of next generation lithium-ion batteries

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

Research Team	Name / Post	Unit / Department / Institution
Principal Investigator	Lu Xiaoying/Assistant Professor	Faculty of Science and Technology/ Technological and Higher Education Institute of Hong Kong (THEi)
Co-Investigator(s)	Yang yong/Professor Fan Jing/Assistant Professor	Department of Chemistry/ College of Chemistry and Chemical Engineering/ Xiamen University /Department of Mechanical Engineering/City College of New York
Others		

3. Project Duration

	Original	Revised	Date of RGC / Institution Approval *(must be quoted)*
Project Start Date	2020/01/01	N.A.	N.A.

Project Completion Date	2022/06/30	2022/12/31	2022/06/21 (by Institution)
Duration *(in month)*	30 months	36 months	2022/06/21
Deadline for Submission of Completion Report	2023/06/30	2023/12/31	2022/06/21

4.4 Please attach photo(s) of acknowledgement of RGC-funded facilities / equipment.

Part B: The Final Report

5. Project Objectives

5.1 Objectives as per original application

1. To devise electrospinning of composite separators with thermally conductive ceramics and plastic waste for micro/nanoscale structure, high crystallinity and compatible interfaces;
2. To characterize and optimize the physiochemical and thermal properties of composite separators for excellent electrochemical performances and superior safety performance;
3. To gain fundamental understandings of process-property-performance relationship by experimental and numerical studies on composite separators for rational design of LIBs

5.2 Revised objectives

Date of approval from the RGC: N.A.

Reasons for the change: N.A.

1.

2.

3. ....

5.3 Realisation of the objectives
(Maximum 1 page; please state how and to what extent the project objectives have been achieved; give reasons for under-achievements and outline attempts to overcome problems, if any)

All the objectives proposed in this project are achieved with the below explanations.

For objective 1, composite separators with thermally conductive ceramics and plastic waste for micro/nanoscale structure, high crystallinity and compatible interfaces was fabricated through optimized electrospinning technology. Thermally conductive metal nitrides were investigated as ceramic fillers for composite separators. PET-based bottles were selected, followed by dissolution in organic solvent. The results revealed that composite separators with micro/nanostructures were successfully synthesized at an applied voltage of 15 kV, distance of 15 cm, needle size of G20, dosing speed of 0.1 mL min^{-1} and solid content of 10 wt.%. After dehydrated condensation reaction, strong chemical bonds between ceramics and polymers were formed. Through systematic study, the objective 1 are successfully achieved.

For objective 2, the composite separators with good electrochemical performances and superior safety properties were well characterized with different tests. The morphology and fiber dimensions were investigated by scanning electron microscope and were controlled by electrospinning process. The crystal phases and crystallinities of separators treated with plasma irradiation were studied by XRD. The compatible interface between polymer and ceramics is advantageous for effective heat dissipation were confirmed by TEM, and FTIR analysis. The separator wettability was evaluated with electrolyte droplet by contact angle measurement in different status. For thermal tests, the thermal stability was estimated in a hot oven by exposing composite separators to 150~250^{o}C for 30 min. The thermal properties including melting and decomposition temperatures was studied with simultaneous thermal analysis. The thermal conductivity of composite separators were measured by laser flash method. Thermal conductivity of composite separators were greatly improved upon the addition of ceramic nitrides, based on parallel model estimation. For electrochemical stability test, composite separators were characterized by LSV. The electrochemical inertness of PET polymer and metal nitrides ensure high electrochemical stability. Ionic conductivity of composite separators were measured by AC impedance spectroscopy with electrochemical station. Ionic conductivity also increased by the relatively large pore size and electrolytephilic property of metal nitrides.

For objective 3, the composite separators process-protperty-performance relationship were well studied in this stage.Comprehensive experimental and numerical simulation are executed to disclose the heat dissipation inside single cells by thermally conductive separators. For this project, composite separators of different thermal conductivities were assembled with identical anodes, cathodes and electrolytes in pouch cell prototypes for ARC measurement. Specifically, prototype cells are charged with 0.1 C to SoC of 100% and placed in adiabatic jackets for ARC experiment. ARC thermal results were compared with DSC results. In addition, ARC test coupled with in-situ electrochemical test are used to evaluate prototype cells under high charge-discharge current densities. The above ARC measurement of the composite separators are conducted in duplicate for reproducibility. Therefore, the relationship between thermal failure temperatures and properties of composite separators can be well established.

Overall, the above experiment and simulation of composite separators are meaningful for building process-property-performance relationship. In this project, composite separators with superior thermal characteristics are developed through achieving these three objectives.

Summary of objectives addressed to date

Objectives *(as per 5.1/5.2 above)*	**Addressed** *(please tick)*	**Percentage Achieved** *(please estimate)*
1. *To develop fluorinated Si/C composites with porous and micro-/nanoarchitectures by nano-bubble-assisted templates followed by spray-pyrolysis to produce a designed structure;*	✓	100%
2. *To characterize and optimize the physical properties of fluorinated Si/C composites to achieve high areal capacity, excellent rate capability and a long life cycle;*	✓	100%
3. *To gain fundamental understandings of the relationship between physical and electrochemical properties;*	✓	100%

6. Research Outcome

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

Preventing short circuit hazard due to the lithium (Li) dendrite formation across the separator from anode of lithium-ion battery (LIBs) throughout operation is important, while conventional separator materials cannot fulfil the increasing safety standard in next generation LIBs. Thus, developing separator materials with high Li dendrite suppression ability in order to prevent from short circuit is of paramount importance for next generation LIBs. In this study, aluminum nitrile modified polyethylene terephthalate (AlN/PET) with micro/nanoarchitecture composites were synthesized with PET which recycled from commercial waste bottle by electrospinning strategy. Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) suggested that AlN nanoparticles was encapsulated in PET micro/nanoarchitecture fibre. Thermogravimetric analysis indicated that the AlN content in composite materials was determined to be about 4-5 wt%. X-ray photoelectron spectroscopy (XPS) and frontier-infrared spectroscopy (FTIR) confirmed that PET polymer structure in PET/AlN composites. N2 adsorption-desorption isotherm showed that PET/AlN 5wt% composite exhibited a porosity of 69.23% and a high electrolyte uptake of 521.69 %. Most importantly, electrochemical results revealed that when evaluated at a current density of 0.5C, PET/AlN 5wt% composites could deliver a reversible specific capacity of 241.7 mAh g-1 after 100 cycles. When C-rate capability tests were conducted at high charge-discharge densities of 0.2, 0.5, 1, 2, and 4C, PET/AlN 5wt% composite manifested average specific capacities of about 246.5, 225.1, 208.2, 179.2, and 116.5 mAh g-1, respectively. The excellent electrochemical performance of PET/AlN 5wt% composite was probably attributed to the combined benefits from AlN nanoparticles, micro/nanoarchitectures and mesoporous structure. These unique features of PET/AlN were advantageous for effective Li ion transport in the repeated charge-discharge cycles and strong hydrothermal stability, thereby resulting in safety, high capacity and excellent C-rate performance. Overall, this study demonstrated the excellent electrochemical performance of PET/AlN composites as stable separator materials for advanced LIBs.

The major findings were already summarized into a manuscript, which is ready for submitted for peer-reviewed journal.

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

Potential for further development of this research is summarized as below:
- The synthesis strategies of separators with superior safe properties developed in this project can be applied to other energy storage materials for performance enhancement of electrochemical processes;
- Theoretical knowledge of separators composites can be further investigated with in-situ electrochemical characterization techniques (e.g. Raman, X-ray diffraction, Fourier-transform infrared spectroscopy etc.) and computational quantum mechanical modelling; and
- The promising results of composite separators can be further extended to investigate the recycling and reuse of similar plastic materials for development of high-performance energy storage materials.

7. Layman's Summary

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

Preventing the risk of short circuits of lithium-ion batteries (LIBs) is crucial. In this study, we synthesized composite materials using recycled polyethylene terephthalate (PET) from commercial waste bottles, which were modified with aluminum nitride (AlN) using an electrospinning technique. Microscopic analysis revealed that the AlN nanoparticles were encapsulated within the PET fibers. Thermal analysis confirmed that the composite materials contained approximately 4-5% AlN by weight. Spectroscopic analysis further confirmed the presence of PET polymer in the PET/AlN composites. Importantly, electrochemical tests showed that the PET/AlN composite with 5% AlN could deliver a reversible specific capacity of 241.7 mAh g^{-1} after 100 cycles when evaluated at a current density of 0.5C. The exceptional electrochemical performance of the PET/AlN composite can be attributed to the combined advantages of AlN nanoparticles, the micro/nanoarchitectures of the PET fibers, and the mesoporous structure of the composite. These unique features facilitate efficient transport of lithium ions during repeated charge-discharge cycles and ensure strong hydrothermal stability, resulting in enhanced safety, high capacity, and excellent performance at different charge-discharge rates. Overall, this study demonstrates that PET/AlN composites can serve as stable separator materials for advanced LIBs, offering outstanding electrochemical performance.

Part C: Research Output

8. Peer-Reviewed Journal Publication(s) Arising Directly From This Research Project

(Please attach a copy of the 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) *(denote the correspond-ing 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 RGC *(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)*						
2022				Jianhua Ge , Zhongfei Liu , Minghui Guan, Juner Kuang , Yuhua Xiao , Yang Yang , Chi Him Tsang , Xiao-Ying Lu *, Chunzhen Yang	Investigation of the electrocatalytic mechanisms of urea oxidation reaction on the surface of transition metal oxides	2022	YES	YES	No
2023				Jiadong Liu, Tsz Yau Mak , Zhe Meng , Xuyang Wang , Yulin Cao , Zhouguang Lu , Dawson Wai-Shun Suen , Xiao-Ying Lu *, Yuanyuan Tang	Efficient recovery of lithium as Li2CO3 and cobalt as Co3O4 from spent lithium-ion batteries after leaching with p-toluene sulfonic acid	2023	YES	YES	No
			✓	Marco Yu Lam Wong , Chi-Wing Tsang, Alpha Chi Him Tsang , Xiao-Ying Lu *	Development of AlN loaded PET separators from waste water bottle plastics with superior thermal characteristics for next generation lithium-ion batteries		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 conference abstract)

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 RGC** *(Yes or No)*	**Accessible from the Institutional Repository** *(Yes or No)*
11/2021 Online	Electrospinning Synthesis of PET-AIN Composite Separators for Advanced Lithium-ion Batteries	The 6th International Conference on New Energy and Future Energy Systems	No	Yes	Yes	No

10. Whether Research Experience And New Knowledge Has Been Transferred / Has Contributed To Teaching And Learning
(Please elaborate)

Three Final year project (FYP) topics were developed for undergraduates of Technological and Higher Education Institute of Hong Kong (THEi). Under the supervision of principle investigator, these students could gain basic knowledge of separator electrospinning synthesis , material characterizations, electrochemical experiments, and battery performance analysis etc.

11. 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**
[redacted]	Environmental Engineering and Management	2017	2021/Graduation
[redacted]	Environmental Engineering and Management	2017	2021/Graduation

[redacted]	Environmental Engineering and Management	2017	2021/Graduation

12. Other Impact

(e.g. award of patents or prizes, collaboration with other research institutions, technology transfer, teaching enhancement, etc.)

PI, Dr. Lu Xiaoying who participated in this project successfully won Best Oral Presentation Award at The 6th International Conference on New Energy and Future Energy Systems, Technology and Innovation (2021) for her excellent performance in the oral presentation.

13. Statistics on Research Outputs

	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	2	1	0	0	Type	No.

14. Public Access Of Completion Report

(Please specify the information, if any, that cannot be provided for public access and give the reasons.)

Information that Cannot Be Provided for Public Access	**Reasons**