RGC Ref. No.:
UGC/FDS25/E10/17
(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 a novel photocatalytic membrane system for bacteria-resistant organics oxidation in wastewater

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

Research Team	Name / Post	Unit / Department / Institution
Principal Investigator	Dr/TSANG Chi Wing	Faculty of Science and Technology/ Technological and Higher Education Institute of Hong Kong
Co-Investigator(s)	Dr Dickson YAN Yuk Shing	The Wasted Co. Ltd. (private business)
Co-Investigator(s)	Prof. LO Man Chi, Irene	Department of Civil and Environmental Engineering/The Hong Kong University of Science and Technology
Co-Investigator(s)	Dr. K. J. An, Alicia	School of School of Energy and Environment/City University of Hong Kong
Co-Investigator(s)	Dr. LAM Leung Yuk, Frank	Department of chemical and Biomolecular engineering/ The Hong Kong University of Science and Technology

3. Project Duration

	Original	Revised	Date of RGC / Institution Approval *(must be quoted)*
Project Start Date	01/01/2018	N/A	N/A
Project Completion Date	31/12/2020	30/6/2021	21/04/2020
Duration *(in month)*	36	42	21/04/2020
Deadline for Submission of Completion Report	31/12/2021	30/6/2022	21/04/2020

Part B: The Final Report

5. Project Objectives

5.1 Objectives as per original application

1. to improve the outdoor water quality by exploring the feasibility of novel visible light driven photocatalysts for antibiotic degradation;
2. to deposit different types of metals onto the surface of photocatalysts to enhance their photoreactivity;
3. to attach the emerging photocatalysts onto a membrane to prevent the nano-sized photocatalysts from discharging into the environment; and
4. to develop a photocatalytic membrane reactor for a continuous treatment of antibiotic containing wastewater.

5.2 Revised objectives

Date of approval from the RGC: N/A

Reasons for the change: N/A

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

Objectives 1 - to improve the outdoor water quality by exploring the feasibility of novel visible light driven photocatalysts for antibiotic degradation.

In view of the photocatalytic potential of bimetallic BiOBr and the limited study for non-TiO_2 photocatalytic membrane reactor (PMR), we developed a PMR with emerging bimetallic BiOBr photocatalyst for the degradation of persistent antibiotics in wastewater under visible light irradiation. It is believed that the development of novel photocatalytic membrane will broaden the horizon of scientific catalytic research and membrane science. In view of engineering application, we have designed a reactor for the future development of pilot-/industrial-scale PMR for antibiotic oxidation and outdoor water quality improvement. The new technology and photocatalytic reactor will significantly improve the outdoor water quality which is a critical issue for urbanized countries and shed light upon the long-term sustainability in environmental research.

Objectives 2 - to deposit different types of metals onto the surface of photocatalysts to enhance their photoreactivity.

Various metal photocatalysts such as Au-BiOBr, Ag-BiOBr and Cu-BiOBr flakes has been fabricated and the efficiency of norfloxacin (a textile dye) degradation by Au-BiOBr, Ag-BiOBr has been studied. Based on the Ag-BiOBr series, we have further developed the Cu-doped BiOBr catalyst series, which further enhance the adsorption and visible-light driven photocatalytic degradation of norfloxacin. The introduction of Cu into BiOBr enhanced the adsorption capacity between the photocatalyst and norfloxacin, which we considered to be the main contribution to its improved performance.

Objectives 3 - to attach the emerging photocatalysts onto a membrane to prevent the nano-sized photocatalysts from discharging into the environment.

The bimetallic Ag-BiOBr and Cu-BiOBr have been attached on alumina membrane successfully, and there is very little leaching of the Ag^{+}, Cu^{2+} and Bi^{3+} from the photocatalysts. After the photodegradation, metal leaching from the catalyst was measured to be 197 ppb and 93 ppb for Cu^{2+} and Bi^{3+}, respectively, which is well below the threshold permitted value for Cu (1 ppm) and Bi (15 ppm) according to the Environmental Protection Agency.

Objectives 4 - to develop a photocatalytic membrane reactor for a continuous treatment of antibiotic containing wastewater.

The photocatalytic membrane reactor for a continuous treatment of antibiotic containing wastewater was developed. The experimental setup consisted of gear pumps (one each for the permeate and feed), feed and permeate containers, liquid conveyance connections, and the direct contact membrane distillation module with hydrophobic membranes for separating the feed and permeate. The hot feed (60 ± 0.5 °C) and cold permeate (20 ± 0.5 °C) were at a flow rate of 30 L h^{-1}. A Cu mesh was used as the electrode and placed over the membrane inside the module and connected to the potentiostat station to measure electrical impedance as shown in **Figure 1**.

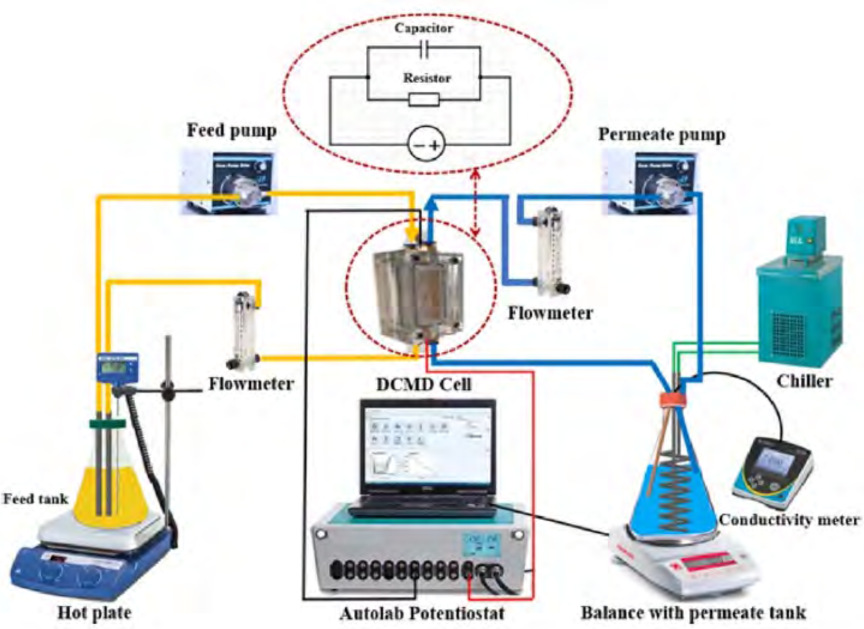

Figure 1. Direct contact membrane distillation setup with a potentiostat for the impedance tests.

5.4 Summary of objectives addressed to date

Objectives *(as per 5.1/5.2 above)*	**Addressed** *(please tick)*	**Percentage Achieved** *(please estimate)*
1. to improve the outdoor water quality by exploring the feasibility of novel visible light driven photocatalysts for antibiotic degradation;	Yes	100%
2. to deposit different types of metals onto the surface of photocatalysts to enhance their photoreactivity;	Yes	100%
3. to attach the emerging photocatalysts onto a membrane to prevent the nano-sized photocatalysts from discharging into the environment; and	Yes	100%
4. to develop a photocatalytic membrane reactor for a continuous treatment of antibiotic containing wastewater.	Yes	100%

6. Research Outcome

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

In this experimental research, an innovative electrospun photocatalyst self-cleaning BiOBr/Ag membrane is introduced for the membrane distillation (MD) treatment of dyeing wastewater coupled with post-MD UV and visible light exposure for fouled membrane regeneration. The E-BiOBr/Ag membrane was fabricated successfully by coating an electrospun membrane with BiOBr/Ag catalyst particles using electrospray technology, with the goal to achieve high hydrophobicity. Along with the E-BiOBr/Ag membrane, two commercial polyvinylidene difluoride (PVDF) and polytetrafluoroethylene (PTFE) membranes were tested for comparison. The fouling processes on all three membranes were monitored in real-time using optical coherence tomography (OCT). The coating of BiOBr/Ag particles on the E-BiOBr/Ag membrane's surface accelerated dye foulant degradation through the electronholes'strong oxidization capacity when exposed to UV. Meanwhile, after Ag nanoparticles were coated on the BiOBr photocatalyst by UV deposition method, not only improved the efficiency of electron separation and transfer but also lessened the electron recombination phenomenon effectively. Correspondingly, compared to the two commercial membranes, the BiOBr/Ag photocatalyst membrane achieved significant improvements in the recovery efficiencies of the water contact angle (95.6%) and water flux (92.2%) under UV illumination, pointing to its potential for fouled membrane regeneration (please refer to Part C of this report: *Chemical Engineering Journal* **2019**, *378*, 122137). In addition, the deposition of Ag on BiOBr as cocatalyst enhanced the visible light harvesting. Finally, the BiOBr/Ag photocatalyst membrane maintained good flux recovery and dye rejection (99.9%) over a 5-cycle MD test coupled with visible light exposure, suggesting the application of the novel self-cleaning photocatalyst membrane as a potential alternative for upscaling MD technology to the industrial level.

Copper is a photoactive transition metal, thus its presence in a photocatalyst may modify the electronic and photophysical properties of the overall material. Thus, we have developed a Cu-doped BiOBr catalyst and fabricated it on membrane using a solvothermal method and assessed for their ability to degrade norfloxacin under visible light. The as-prepared Cu-doped BiOBr showed activity superior to BiOBr in the photocatalytic degradation of norfloxacin under visible-light irradiation, which was attributed to its improved light-harvesting properties, enhanced charge separation and interfacial charge transfer. Furthermore, we found for the first time that the introduction of Cu into BiOBr enhanced the adsorption capacity between the photocatalyst and norfloxacin, which we considered to be the main contribution to its improved performance. Cu-doped BiOBr containing the optimal proportion of Cu and Bi (Cu:Bi = 0.03) had a photocatalytic degradation constant of $0.64 \times 10{-}2\ \mathrm{min}^{-1}$, which is 2.28 times higher than that of undoped BiOBr. The primary oxidation pathway was determined to involve the transfer of photogenerated holes to norfloxacin. Finally, we demonstrated that the Cu-doped BiOBr photocatalyst retained 95% of its initial activity even after five successive catalytic cycles, confirming its excellent recyclability (please refer to Part C of this report: *Chemical Engineering Journal* **2020**, *401*, 126021).

Apart from fouling problem in membrane distillation, which can be solved by self-cleaning method, wetting of hydrophobic membranes is considered to be one of the major limitations that must be overcome to further the development of membrane distillation technology. Low surface tension liquids can induce wetting, which can significantly affect permeate conductivity after the complete wetting of the membrane pores. A thin conductive hydrophobic polyaniline (PANI) layer was sandwiched between two nonconductive polyvinylidene fluoride (PVDF) layers by the phase inversion method. The surface of this PANI-PVDF membrane was then electrosprayed with polytetrafluoroethylene to impart superhydrophobicity (water contact angle, ∼160°). Finally, the wetting of membranes was monitored and detected during direct contact membrane distillation of a low surface tension saline feed containing sodium dodecyl sulfate by electrochemical impedance spectroscopy. Compared with measuring conductivity of the permeate side, we found that measuring the cross-membrane impedance at a constant frequency (100 kHz) demonstrated more precise detection and a superior ability in distinguishing different stages of wetting and their intrusion. Furthermore, our experiments demonstrated the possible strategies to elude membrane wetting by flushing distilled water periodically (please refer to Part C of this report: *ACS Applied Polymer Materials* **2021**, *3*, 679-690).

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

1. It could be interesting to use process modeling to study the techno-economic aspects of the membrane distillation technology, using the as-developed photocatalytic method. This could be done using chemical process modeling software such as SuperPro Designer® or ASPEN Plus to determine the economics of the water purification process. After knowing the different synthetic cost of the catalyst and the membrane, the net present value and internal rate of return could be estimated and the potentials of the economic value of the process can be determined.

2. To demonstrate the activation of the BiOBr/Ag photocatalyst membrane under visible light, a 5-cycle MD test was conducted, each round consisting of a 8 h membrane distillation operation followed by 2 h visible light illumination for membrane regeneration. the BiOBr/Ag photocatalyst can be regenerated over 90% after fifth round of the membrane distillation test, pointing to its effectiveness in significantly extending the lifespan of the membrane and, consequently, its potential for accelerating membrane distillation's commercialization. Thus, wastewater treatment company could potentially consider the current developed technology for further commercialization.

7. Layman's Summary

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

Conventional wastewater treatment plants which depend heavily on biological treatments are considered ineffective to remove pharmaceuticals, dyes, etc. One of the popular alternatives is advanced oxidation processes. By exciting photocatalyst with various light sources, non-selective hydroxyl radicals can be generated to oxidize most of the complex organics and to mineralize them into harmless inorganic molecules. TiO_2 is the most commonly used photocatalyst which has been studied over decades. However, one of the drawbacks is that it can only be excited under UV irradiation and thus, an additional cost of illumination is needed for wastewater treatment. In order to utilize visible light source, novel photocatalysts were developed in which bismuth oxybromide (BiOBr) was doped with various photoactive elements such as Ag and Cu and they showed a very high visible-light response and a stable structure under irradiation. The Ag-BiOBr and Cu-BiOBr were successfully deposited on to the surface of alumina membrane and after photodegradation, metal leaching from the catalyst was measured to be well below the threshold permitted value for Cu^{2+} and Bi^{3+}, respectively, according to the Environmental Protection Agency. The organic waste molecules in the wastewater were successfully degraded on the membrane surface, whereas clean water passed through the membrane.

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)*						
2019	N/A	N/A	N/A	Jiaxin Guoa, Dickson Y.S. Yan, Frank L.Y. Lam, Bhaskar Jyoti Deka, Xincong Lyu, Yun Hau Ng, and Alicia Kyoungjin An	Self-cleaning BiOBr/Ag photocatalytic membrane for membrane regeneration under visible light in membrane distillation / *Chemical Engineering Journal* **2019**, *378*, 122137.	No	Yes (see Annex 1)	Yes	Yes
2020	N/A	N/A	N/A	Xincong Lv, Dickson Y.S. Yan, Frank Leung-Yuk Lam, Yun Hau Ng, Shengming Yin, Alicia Kyoungjin An	Solvothermal synthesis of copper-doped BiOBr microflowers with enhanced adsorption and visible-light driven photocatalytic degradation of norfloxacin/ *Chemical Engineering Journal* **2020**, *401*, 126021.	No	Yes (see Annex 2)	Yes	Yes
					A Conductive Hydrophobic				

2021	N/A	N/A	N/A	Bhaskar Jyoti Deka, Jiaxin Guo, Pak Wai Won, Noman Khalid Khanzada, Jehad A. Kharraz, Chi-Wing Tsang, and Alicia Kyoungjin An	Polyaniline Sandwiched Polyvinylidene Fluoride Membrane for Early Detection of Surfactant-Induced Wetting in Membrane Distillation Using Impedance/ *ACS Applied Polymer Materials* **2021**, *3*, 679-690.	No	Yes (see Annex 3)	Yes	Yes
2021	N/A	N/A	N/A	Subramania, K.; Sarkar, M. K.; Wang, H.; Leu, S.-Y.; Tsang, C.-W.; Chopra, S. S.; Jin, M.; Nallapaneni, M. K.; Kumar, V.; Chan, E. M. H.; Lu, X.-Y.; Chen, C.; Li, X.; Lin, C. S. K.	An overview of cotton and polyester, and their blended waste textile valorisation to value-added products: A circular economy approach – research trends, opportunities and challenges/ *Critical Reviews in Environmental Science and Technology* **2021** DOI:10.1080/10643389.2021.1966254.	No	Yes (see Annex 4)	Yes	Yes

(a) Jiaxin Guo, Bhaskar Deka, Shengming Yin, Pak Wai Wong, Noman Khanzada, Jehad Kharraz and Xincong Lv are research assistants of Alicia An's research group, responsible for catalyst preparation and catalytic performance evaluation.
(b) Dr Yun Hau Ng is associate professor at City University of Hong Kong and Frank Lam is assistant professor at Hong Kong University of Science and Technology.
(c) Karpagam Subramanian, Huaimin Wang, Zi-Hao Qin, Mushan Jin, Chao Chen are research assistants of Dr Carol Lin's research group, responsible for literature review and manuscript preparation.
(d) Dr Shauhrat S. Chopra and Dr Carol Lin are assistant and associate professor at City

University of Hong Kong, respectively.
(e) Dr Vinod Kumar is senior lecturer at Cranfield University.

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)*
15-17 Nov 2020/Webinar	"Textile wastewater treatment using a new type of BiOBr-derived photocatalytic membrane: A comparison with current state-of-the-art filtration technology"	*International Conference for Materials Engineering and Nanotechnology 2020,* Shenzhen	No	Yes (see Annex 5)	Yes	No

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

1. Shared new findings to undergraduate degree students at Technological and Higher Education Institute of Hong Kong on waste water treatment modules.
2. Given talks at the research seminar held at THEi.

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]	[redacted]	[redacted]	[redacted]

* This student's thesis title page is attached in this report.

In this project, I have trained 1 undergraduate student:

[redacted]

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

1. Collaborated with Dr. Carol Sze Ki Lin from City University of Hong Kong on

textile waste valorisation and textile wastewater treatment (*Critical Reviews in Environmental Science and Technology* **2021** DOI:10.1080/10643389.2021.1966254.).

2. THEi Researcher Award 2018
3. Invited as a Session Chair at the *International Conference for Materials Engineering and Nanotechnology 2020 (Please see attached Certificate of being the Session Chair)*
4. Invited lectures to master degree students at City University of Hong Kong on waste treatment topics.

13. Statistics on Research Outputs

	Peer-reviewed Journal Publications	Conference Papers	Scholarly Books, Monographs and Chapters	Patents Awarded	Other Research Outputs (please specify)	
					Type	No.
No. of outputs arising directly from this research project	4	1	N/A	N/A	N/A	N/A

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
N/A	N/A