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(please insert ref. above)

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

Investigation of new multi-functional materials based on hierarchical porous mixed oxides and carbon aerogels for air purification and disinfection

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

	Hong Kong Team	Mainland Team
Name of Principal Investigator *(with title)*	Prof. YEUNG King Lun	Prof. QIU Jieshan
Post	Full Professor	Cheung Kong Professor
Unit / Department / Institution	Department Chemical and Biological Engineering, Division of Environment and Sustainability / The Hong Kong University of Science and Technology	Faculty of Chemical, Environmental and Biological Science and Technology / Dalian University of Technology
Contact Information	kekyeung@ust.hk	jqiu@dlut.edu.cn
Co-investigator(s) *(with title and institution)*	Prof. KWAN Joseph KC, Health, Safety and Environmental Office; Dr. HAN Wei, Division of Environment	Prof. MO Ziyao, Guangzhou State Key Laboratory of Respiratory Disease

3. Project Duration

	Original	Revised	Date of RGC/ Institution Approval *(must be quoted*)
Project Start date	January 1, 2014		
Project Completion date	December 31, 2017		
Duration *(in month)*	48		

Deadline for Submission of Completion Report	December 31, 2018		

Part B: The Completion Report

5. Project Objectives

5.1 Objectives as per original application

1. To design, synthesize and characterize mixed oxides and carbon aerogels with hierarchical pore structures for molecular and particulate filtrations;

2. To incorporate active functional sites for (i) capture of airborne microorganisms, (ii) adsorption of molecular pollutants, (iii) catalytic conversion of molecular pollutants, (iv) inactivation of microbial contaminants;

3. To observe in operando the molecular mechanism and process of capture, transformation and inactivation of pollutants in these materials under practical use conditions;

4. To carry out material and process optimization using mathematical modeling based on experimental evidence for the design of a prototype for field test.

5.2 Revised Objectives

Date of approval from the RGC: _____

Reasons for the change: _____

_____

1.
2.
3. ....

6. Research Outcome

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

1. Aerogel matrices

Silica and silica-titania aerogels were prepared from silicon and titanium alkoxides via two-step sol-gel process (i.e., acid-catalyzing hydrolysis and base-catalyzing condensation) and supercritical drying process (*Ref. 9-Poster 9*). Freestanding non-silica, metal oxide aerogels were also prepared using gelation agents such as propylene oxides. These metal oxide aerogels can be transformed into MOFs aerogel using a new synthesis technique developed in the laboratory (*Ref. 8-Paper 3; 9-Oral 5; 9-Poster 2, 8 & 10*). Graphene aerogels (or cryogels) were prepared by using amine-containing cross linkers to assemble graphene oxide suspension during hydrothermal process, followed by supercritical (or freeze) drying process (*Ref. 9-Oral 3*).

2. Active material incorporation

New preparation methods were developed to spatially incorporate active functional sites (e.g., metals, metal oxides, metal complexes) into different locations (e.g., external surface, interlayer, core) of nanotubes by adjusting type, addition sequence, surface charge and concentration of active precursors (*Ref. 11-Patent 1*). A series of high active

adsorbents (e.g., zeolites, Fe/zeolites, metal-organic framework materials) and catalysts (e.g., VO_x/TiO_2, $Cu/VO_x/TiO_2$, $Au/VO_x/TiO_2$, CeO_2) were developed for effective capture and conversion of molecule pollutants including VOCs and malodors (mainly NH_3 and H_2S) (*Ref. 9-Oral 4, 7; 9-Poster 5, 6*). The above-mentioned active materials were incorporated into oxide and graphene aerogels by ex-situ (i.e., assembling active materials during aerogel preparation) and in-situ (i.e., growing active materials from precursors during aerogel preparation) routes (*Ref. 9-Oral 1, 6; 9-Poster 1, 3, 4, 7*). The obtained active oxide and graphene aerogels exhibited excellent adsorption and catalytic conversion performances for molecule pollutants and enhanced filtration efficiencies for particulate matter as well as bactericidal activities (*Ref. 8-Paper 4; 9-Oral 3*).

3. Mechanism investigation

Operando Raman-online FTIR system was used to investigate catalytic mechanisms for molecule pollutants (e.g., toluene, NH_3). The nature of the active sites, configurational adsorption of toluene and the reactive oxygen species play important roles in the catalyst activity and selectivity (*Ref. 8-Paper 1, 2; 9-Oral 2*). The reaction intermediate of NH_3 selective oxidation on VO_x/TiO_2 catalyst is hydrazinium ion. The proposed reaction route is NH_3 adsorption, the formation and decomposition of hydrazinium ion to N_2 and H_2O (*Ref. 9-Oral 7; 9-Poster 9*).

4. Material optimization

Microporous material including zeolites and metal-organic framework materials were incorporated into oxide and graphene aerogel matrixes to generate optimally designed hierarchical porous structures (*Ref. 9-Poster 3, 4, 7*). Bimetal-organic framework materials were transformed into low-dimensional mixed metal oxides by controlled thermal decomposition process. The optimized metal oxides were capable of effectively converting molecule pollutants under near-ambient conditions (*Ref. 11-Patent 2*).

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

Research outcomes have led to a grant from the Guangzhou Collaborative Innovation Key Program. We are cooperating with a mainland company to scale up oxide aerogels for practical application of VOC catalytic conversion under near-ambient conditions.

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)

This project successfully developed new synthesis routes for preparing aerogels from diverse materials including metal oxides, metal-organic frameworks and graphene that has unique mechanical, thermal, electrical and transport properties. Studies show that aerogels particularly graphene aerogels can filter particulates from air as good as the European H11 HEPA filters. It can simultaneously capture and inactivate airborne bacteria. Furthermore, graphene aerogel can adsorb toluene, an important volatile organic pollutant found indoor. Incorporation of catalysts allow the graphene aerogel to treat VOCs such as acetone, methanol, cyclohexane and toluene.

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)*						
2018				**Qingyue Wang**, **King Lun Yeung***, Miguel A. Bañares*	Operando Raman-online FTIR investigation of ceria, vanadia/ceria and gold/ceria catalysts for toluene elimination, Journal of Catalysis, 364: 80-88	December 31, 2018	Yes	Yes	Yes
			√	**Qingyue Wang**, **King Lun Yeung***, Miguel A. Bañares*	A Review of Ceria Catalysts for VOCs Remediation (draft completed to be submitted to Cat. Today)	December 31, 2018	Yes	Yes	No
			√	**Zhang Liu**, **Wei Han**, **King Lun Yeung***	A facile conversion route for metal organic framework (MOF) aerogels (draft completed to be submitted to Angew. Chemie)	December 31, 2018	Yes	Yes	No

			√	**Weiyang Chen**, **King Lun Yeung***	Catalytic graphene aerogel for air purification and disinfection (draft completed to be submitted to Appl. Catal. B)	December 31, 2018	Yes	Yes	No

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)*
08/2018/ Boston, USA	Ultralight, monolithic graphene and ceria aerogels for VOCs removal and disinfection (Oral 1)	256th ACS National Meeting & Exposition	December 31, 2018	Yes	Yes	Yes
04/2017/ San Francisco, USA	Operando Raman-FTIR study of gold catalysts supported on titania and ceria for VOC-related odor elimination (Oral 2)	253rd ACS National Meeting & Exposition	December 31, 2018	Yes	Yes	Yes
08/2016/ Philadelphia, USA	Characterizations of the transport properties of graphene aerogels: electrical, thermal, and gas transport properties (Oral 3)	252nd ACS National Meeting & Exposition	December 31, 2018	Yes	Yes	Yes

11/2015/ Salt Lake City, USA	Synthesis, characterization of $Au/V_2O_5/CeO_2$, $Au/V_2O_5/TiO_2$ catalysts and their performance in VOC-related malodor treatment (Oral 4)	2015 AIChE Annual Meeting	December 31, 2015	No	Yes	Yes
08/2015/ Boston, USA	CPO-27-Ni aerogel incorporated in nickel foam for efficient CO_2 capture (Oral 5)	250th ACS National Meeting & Exposition	December 31, 2015	No	Yes	Yes
06/2015 Singapore	Freestanding porous mixed oxides and graphene aerogels for air purification and disinfection (Oral 6)	8th International Conference on Materials for Advanced Technologies	December 31, 2015	No	Yes	Yes
11/2014/ Atlanta, USA	Synthesis, characterization of vanadia/titania-based catalysts and their performance in ammonia remediation (Oral 7)	2014 AIChE Annual Meeting	December 31, 2015	No	Yes	Yes
08/2014/ San Francisco, USA	Malodor treatment using controlled-release antimicrobial gel (Oral 8)	248^{th} ACS National Meeting and Exposition	December 31, 2015	No	Yes	Yes
04/2017/ San Francisco, USA	Monolithic aerogels of graphene-mixed metal oxides for air purification and disinfection (Poster 1)	253rd ACS National Meeting & Exposition	December 31, 2018	Yes	Yes	Yes
11/2016/ San Francisco, USA	Compressive mechanical properties of metal-organic framework (MOF) aerogels prepared by supercritical drying (Poster 2)	2016 AIChE Annual Meeting	December 31, 2018	Yes	Yes	Yes

11/2016/ San Francisco, USA	Preparation of graphene-MOFs composite aerogels (Poster 3)	2016 AIChE Annual Meeting	December 31, 2018	Yes	Yes	Yes
08/2016/ Philadelphia, USA	Synthesis of MOF-graphene hybrid aerogels (Poster 4)	252nd ACS National Meeting & Exposition	December 31, 2018	Yes	Yes	Yes
08/2016/ Philadelphia, USA	Low temperature catalyst for VOC abatement (Poster 5)	252nd ACS National Meeting & Exposition	December 31, 2018	Yes	Yes	Yes
11/2015/ Salt Lake City, USA	Catalysts for treating H_2S malodor problem at ambient temperature (Poster 6)	2015 AIChE Annual Meeting	December 31, 2015	No	Yes	Yes
11/2015/ Salt Lake City, USA	Design, synthesis and fabrication of freestanding aerogels of graphene-MOFs for air treatment (Poster 7)	2015 AIChE Annual Meeting	December 31, 2015	No	Yes	Yes
11/2015/ Salt Lake City, USA	Preparation, characterization and catalytic performance of vanadium oxide supported on titania nanobundle for NH_3 degradation (Poster 8)	2015 AIChE Annual Meeting	December 31, 2015	No	Yes	Yes
11/2015/ Salt Lake City, USA	Copper/vanadia/titania bifunctional catalysts for ammonia remediation (Poster 9)	2015 AIChE Annual Meeting	December 31, 2015	No	Yes	Yes
08/2015/ Boston, USA	The synthesis of freestanding metal-organic-framework (MOF) aerogels (Poster 10)	250th ACS National Meeting & Exposition	December 31, 2015	No	Yes	Yes
08/2015/ Boston, USA	Aerogel catalysts for direct remediation of NH_3 malodor in air (Poster 11)	250th ACS National Meeting & Exposition	December 31, 2015	No	Yes	Yes

11/2014/ Atlanta, USA	A general route to prepare metal-organic-framework aerogel (Poster 12)	2014 AIChE Annual Meeting	December 31, 2015	No	Yes	Yes
08/2014/San Francisco, USA	Room temperature VO_x/TiO_2 catalyst for treatment of H_2S malodor in air (Poster 13)	248th ACS National Meeting & Exposition	December 31, 2015	No	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
LAI Yue Tak	MPhil	September, 2012	August, 2014
CHEUNG Kei Bo	MPhil	September, 2012	January, 2015
CHANG Qing	PhD	September, 2010	February, 2015
LIU Zhang	PhD	February, 2013	January, 2018
WANG Qingyue	PhD	September, 2014	November, 2018

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

Patents

(1) King Lun Yeung, Shammi Akter Ferdousi and Wei Han, "Incorporating metals, metal oxides and compounds on the inner and outer surfaces of nanotubes and between the walls of the nanotubes and preparation thereof", Chinese Patent (Patent No. ZL201380015460.5), Hong Kong Patent (Patent No. HK1202518), US Patent (Pub. No. US20150050494A1).

(2) King Lun Yeung, Qingyue Wang, Zhimin Li and Wei Han, "Two-dimensional catalytic materials derived from metal-organic frameworks for VOC remediation", US Provisional Patent (Appl. No. US62764371).

Collaboration

(1) Establish the Collaboration on operando technologies with Prof. Miguel A. Bañares' group in Institute for Catalysis and Petrochemistry, Spanish National Research Council.

(2) Establish the collaboration on air treatment technologies with Prof. Suojiang Zhang's group in Institute of Process Engineering, Chinese Academy of Sciences.