



THE UNIVERSITY OF HONG KONG

Theme-Based Research Scheme (TBRS)

on

Challenges in Organic Photo-Voltaics

and Light Emitting Diodes

– A Concerted Multi-Disciplinary

and Multi-Institutional Effort

T23-713/11

Completion Report

December 2017

**RESEARCH GRANTS COUNCIL
THEME-BASED RESEARCH SCHEME (TRS)**

Completion Report on Funded Project

Project start date: 01 January 2012
Project completion date: 31 December 2016

1. Project Title: Challenges in Organic Photo-Voltaics and Light Emitting Diodes – A Concerted Multi-Disciplinary and Multi-Institutional Effort

2. Names and Academic Affiliations of Project Team Members[#]

Project team member	Name / Post	Unit / Department / Institution	Average number of hours per week spent on this project in the <u>whole</u> project period
Project Coordinator (PC)	V.W.-W. Yam / Chair Professor	Chemistry / HKU	3
Co-Principal Investigator(s)	V.W.-W. Yam / Chair Professor	Chemistry / HKU	12
	C.-M. Che / Chair Professor	Chemistry / HKU	10
	H.-S. Kwok / Chair Professor	Electronic & Computer Engineering / HKUST	10
	C.-S. Lee / Chair Professor ¹	Chemistry / CityU	8
	K.-M. Ng / Chair Professor	Chemical & Biomolecular Engineering / HKUST	10
	C. Surya / Endowed Professor	Electronic & Information Engineering / PolyU	10
	F.-R. Zhu / Professor	Physics / HKBU	10
Co-Investigator(s)	W.-K. Chan / Professor	Chemistry / HKU	5
	G.-H. Chen / Professor	Chemistry / HKU	2
	A. Rogach / Chair Professor ²	Materials Science &	3

		Engineering / CityU	
	S.-K. So / Professor	Physics / HKBU	5
	B.-Z. Tang / Chair Professor	Chemistry / HKUST	3
	W.-Y. Wong / Chair Professor ³	Applied Biology & Chemistry Technology / PolyU	6
	M. Wong / Professor	Electronic & Computer Engineering / HKUST	2
	R.M.-S. Wong / Professor	Chemistry / HKBU	5
Collaborators	P.K.-K. Ho / Director	Nano & Advanced Materials Institute Limited	N.A.
	M.H.-W. Lam / Professor	Chemistry / CityU	N.A.
	K.-Y. Wong / Chair Professor	Applied Biology & Chemistry Technology / PolyU	N.A.
	R.A.L. Vellaisamy / Associate Professor ⁴	Materials Science & Engineering / CityU	N.A.
	F. Yan / Professor	Applied Physics / PolyU	N.A.

Please highlight the approved changes in the project team composition and quote the date when the RGC granted approval of such changes. For changes in the project team composition, please submit a separate request, together with the justification and the curriculum vitae of the new member(s), to the RGC three months prior to the intended effective date of the change. N.A.

¹ Prof. C.-S. Lee has now changed to work from Department of Physics and Materials Science at City University of Hong Kong to the Department of Chemistry at City University of Hong Kong starting from 1 January 2017.

² The "Department of Physics and Materials Science" at City University of Hong Kong has been retitled and divided into two Departments, namely "Department of Physics" and "Department of Materials Science and Engineering" starting from 1 July 2017. Prof. A. Rogach is now working in the Department of Materials Science and Engineering at City University of Hong Kong.

³ Prof. W.-Y. Wong has now changed to work from the Department of Chemistry at Hong Kong Baptist University to the Department of Applied Biology and Chemical Technology at The Hong Kong Polytechnic University starting from 16 June 2016.

⁴ The "Department of Physics and Materials Science" at City University of Hong Kong has been retitled and divided into two Departments, namely "Department of Physics" and "Department of Materials Science and Engineering" starting from 1 July 2017. Dr. R.A.L. Vellaisamy is now working in the Department of Materials Science and Engineering at City University of Hong Kong.

3. Project Objectives

Summary of objectives addressed/achieved:

Objectives*	Percentage achieved	Remarks**
1. To develop libraries of patentable robust classes of phosphorescent materials of precious metals, inexpensive metals and/or metals of large natural abundance in China and metal-free small molecules and polymers for OLED, PLED and OPV applications	100 %	Please refer to Section 6.1.
2. To develop highly efficient and new synthetic methods and low-cost high-purity separation processes for solution-processable OPV and OLED materials	100 %	Please refer to Section 6.1.
3. To understand the controlling factors and to develop interfacial engineering methods for enhancing the	100 %	Please refer to Section 6.1.

Objectives*	Percentage achieved	Remarks**
stability and performance of OPVs and OLEDs		
4. To develop patentable manufacturable technology for active matrix organic light emitting diodes (AMOLEDs)	95 %	Please refer to Section 6.1.
5. To develop printing technology platform for large area OPV and OLED devices	95 %	Please refer to Section 6.1.

* *Please highlight the approved changes in objectives and quote the date when the RGC granted approval of such changes.*

** *Please provide reasons for significantly slower rate of progress than originally planned.*

6. Research Highlights and Outputs

(Maximum 20 A4 pages for sections 6 to 10, excluding any appendices and attachments)

6.1 What are the most exciting research accomplishments of the project?

Overview: With the efforts and collaboration contributed by the team members, exciting research accomplishments have been made to address the grand challenges of the project, along with a total of 253 publications in international SCI journals with high impact factors, including *Energy Environ. Sci.*, *Mater. Today*, *Adv. Mater.*, *Adv. Energy Mater.*, *J. Am. Chem. Soc.*, *ACS Nano*, *Adv. Funct. Mater.* and *Angew. Chem. Int. Ed.* Selected highlights of the research accomplishments of the project are summarized as follows.

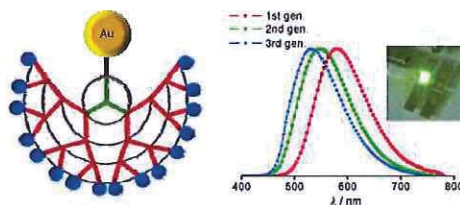
1. Development of OLED Materials

With the efforts dedicated by the team, exceptional performance has been demonstrated on the development of new classes of high-performance OLED materials, especially gold(III) and platinum(II) systems. Various new classes of solution-processable and vacuum-deposited OLED materials have been generated, and highly efficient OLED devices with specifications meeting the industrial standards, including high external quantum efficiencies (EQEs), extremely small efficiency roll-offs, and long operational lifetimes, have been realized. In addition, the team has demonstrated the first report on the utilization of exciplex host for allowing thermally activated delayed fluorescence (TADF) emission. The superior performance has attracted lots of interests from industry. Strong collaborative links with renowned industrial partners.

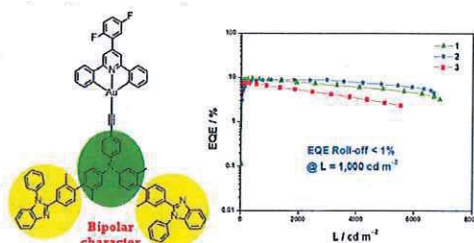
A “TCL-HKU Joint Laboratory for New Printable OLED Materials and Technology” has been established with the funding support from TCL Corporation.

Below are listed the selected outputs related to the deliverables of the project:

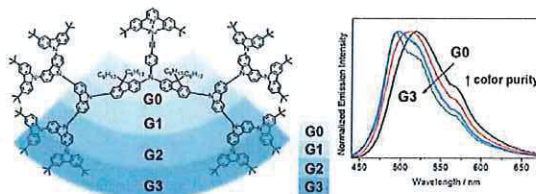
- Successful demonstration on the design and synthesis of phosphorescent dendrimers by the team has opened up a new direction for the preparation of solution-processable phosphorescent OLEDs (PHOLEDs). Apart from the iridium(III), ruthenium(II), and rhenium(I) systems, there have been no reports on the synthesis of dendrimers with other heavy metal centres for the fabrication of PHOLEDs. A novel class of carbazole-based dendritic alkynylgold(III) complexes has been synthesized and demonstrated as phosphorescent emitters in solution-processable OLEDs. The energy as well as the bathochromic shift of the emissions can be tuned effectively through a control of the dendrimer generation. Together with high photoluminescence quantum yield (PLQYs) of up to 0.60 in the doped thin films, superior device performances with high current and EQEs of up to 24.0 cd A^{-1} and 7.8 %, respectively, have been achieved for the optimized device. More importantly, a saturated yellow emission with CIE coordinates of (0.49, 0.50) has been obtained. ***This is the first report on luminescent gold(III) dendrimers*** and represents a new class of dendritic complexes as promising candidates for highly efficient solution-processable PHOLEDs. ***This finding has been reported and published in the Special Issue in celebration of***



- Further extension of this work by utilizing a rigid dendritic structure coordinated to a gold center has led to another class of gold(III)-based bipolar materials containing electron-donating triphenylamine and electron-accepting benzimidazole moieties, which has been applied as phosphorescent dopants in the fabrication of solution-processable PHOLEDs. The incorporation of methyl groups in the central phenyl unit has been found to weaken the π -conjugation between donor and acceptor pairs and rigidify the molecule to reduce non-radiative decay, yielding high PLQYs of up to 0.75 in spin-coated thin films. In addition, highly efficient solution-processable PHOLEDs with EQEs of up to 10.0 % and current efficiencies of up to 33.6 cd A^{-1} have been achieved; particularly, the optimized devices demonstrate superior electroluminescence performance with small EQE roll-off values of less than 1 % and 21 % at practical brightness level of 1,000 and 5,000 cd m^{-2} , respectively. **This is the first example of bipolar alkynylgold(III) complexes in the fabrication of OLEDs with an extremely small EQE roll-off of less than 1 % even at 1,000 cd m^{-2} .** These findings have been published in *J. Am. Chem. Soc.* **2014**, 136, 17861–17868.

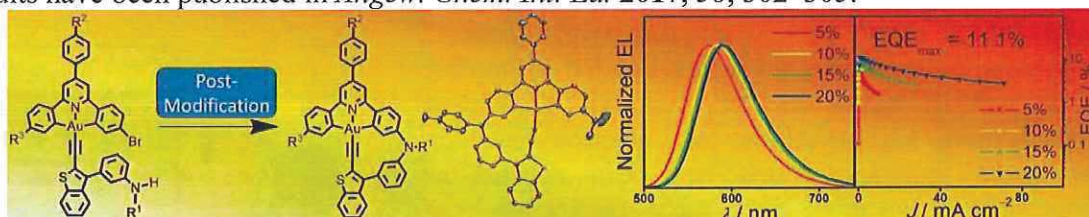


- The development of blue-emitting PHOLEDs is indispensable for full-color displays and solid-state illumination. However, blue-emitting PHOLEDs are still generally considered to be lagging behind in their performance in terms of efficiencies, color purity, and device stability when compared to their red- and green-emitting counterparts. With the aim of tuning emission energies to the blue region, the team has devoted the efforts to design a novel class of blue-emitting alkynylgold(III) dendrimers based on bulky tridentate ligand as dendrons and carbazole moieties as peripheral groups. With the incorporation of bulky carbazole moieties to form higher generation dendrimers, the undesirable excimeric emission could be effectively reduced, allowing the fine-tuning of the emission color toward the blue region. Vibronic-structured emission bands can be clearly resolved with increasing dendrimer generation of up to 3, even at high dopant concentration of 50 wt%. High-performance sky-blue-emitting OLEDs with current efficiencies of up to 23.7 cd A^{-1} and EQEs of up to 6.9 % have been realized by a simple spin-coating technique. This represents the first successful demonstration of sky-blue-emitting alkynylgold(III) complexes and its application in solution-processable OLEDs. This finding also **opens up a new avenue to tackle the biggest challenges for the design and synthesis of sky-blue emitters of square-planar metal complexes of d^8 electronic configuration** and has been published in *J. Am. Chem. Soc.* **2017**, 139, 10539–10550.

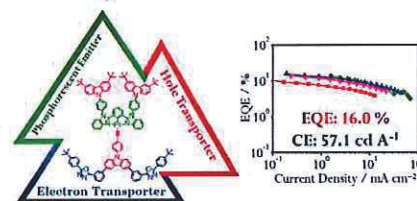


- These successful demonstrations of highly luminescent tridentate-containing gold(III) complexes have inspired the team to generate another new class of cyclometalated tetradentate alkynylgold(III) complexes by post-synthetic modification. Through the judicious design and choice of pincer ligands, post-synthetic cyclization could be achieved to produce the robust and structurally rigid class of tetradentate gold(III) $C^N^C^C$ complexes with high PLQYs of up to 0.49 in solution and 0.78 in doped thin films at room temperature, at least an order of magnitude higher than those of the structurally related uncyclized tridentate alkynylgold(III) analogues. High-performance yellow to orange-red-emitting solution-processable OLEDs have also been achieved with EQEs of up to 11.1 %. These results demonstrate that the intramolecular cyclization can effectively rigidify the molecule and reduce the non-radiative decay. **This work describes for the first time of the use of post-synthetic ligand modification approach to overcome the synthetic challenge for tetradentate alkynylgold(III) complexes**, as well as opens up a new

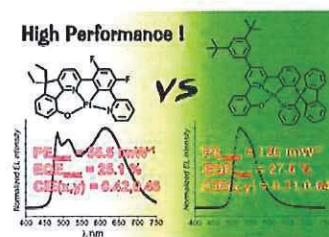
avenue for synthesizing multi-dentate metal complexes and advanced functional organometallic and coordinate compounds, which cannot be accessible by other conventional methods. These results have been published in *Angew. Chem. Int. Ed.* **2017**, *56*, 302–305.



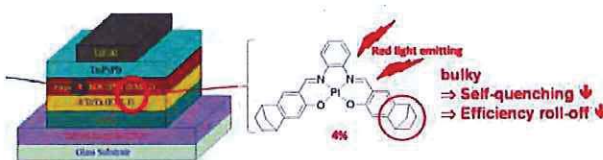
- On the basis of the design strategy of gold(III) dendrimers, the team has combined its efforts to generate a new class of luminescent dendritic carbazole-containing alkynylplatinum(II) complexes. Through the incorporation of a delicate balance of electron-donating carbazole moieties and electron-accepting phenylbenzimidazole or oxadiazole moieties into the platinum(II) core, the platinum(II) dendritic complexes have been demonstrated to exhibit bipolar charge transport character with high PLQYs of up to 0.75 in thin films. It is further found that the *meta*-substitution can effectively suppress the donor-acceptor interaction and thus enhances the bipolar character of the metal complexes. More importantly, high-performance solution-processable green-emitting PHOLEDs with current efficiencies of up to 57.4 cd A⁻¹ and EQEs of up to 16.0 % have been realized. **This EQE is one of the highest values among the reported solution-processable PHOLEDs based on platinum(II) complexes** and is comparable to the state-of-the-art solution-processable OLEDs based on iridium(III) complexes. This finding provides a useful strategy in the design for highly efficient PHOLED applications and has been published in *J. Am. Chem. Soc.* **2017**, *139*, 6351–6362.



- In addition to the development of efficient luminescent alkynylgold(III) system, the team is one of the pioneers in the design and synthesis of platinum(II)-based phosphorescent materials for OLED applications. With the efforts dedicated by the team, a new class of platinum(II)-based complexes with rigid tetradentate O⁺C⁻N⁺C⁻ ligands, which shows strong green emission with high PLQYs of up to 0.95 and good thermal stability with high decomposition temperature of > 423 °C, has been synthesized. Three of the complexes exhibit self-quenching rate constants of 8.5 × 10⁶ to 2.0 × 10⁷ mol⁻¹ dm³ s⁻¹, which are lower than those of other reported platinum(II) complexes. **The optimized yellow- and green-emitting devices based on the complexes exhibit extremely high maximum power efficiencies of 118 and 126 lm W⁻¹, respectively; particularly, these power efficiency values are the highest among the reported platinum(II)-based OLEDs** and are comparable to those of the reported iridium(III) OLEDs without any out-coupling technique. Through the modification of the chemical structure to reduce the intermolecular interactions between molecules, excimer emission can also be successfully suppressed, in which no significant shift in CIE coordinates in the electroluminescence spectra is observed. These findings have been published in *Chem. Sci.* **2014**, *5*, 4819–4830.

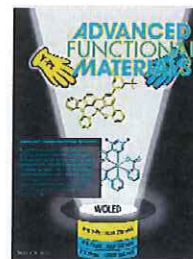


- Further extension of this work by utilizing another O⁻N⁺N⁺O⁻ tetradentate ligand coordinated to the platinum center has generated another new class of sterically hindered platinum(II) Schiff-base complexes. One of the complexes displays intense red emission with PLQY of 0.29 in thin film and exhibits a self-quenching rate constant of 1.0 × 10⁷ mol⁻¹ dm³ s⁻¹. Multilayer PHOLEDs based on these platinum(II) complexes co-doped with a wide band-gap iridium(III) complex as a deep electron trapper have also been prepared. Efficient red-emitting PHOLEDs with maximum current and power efficiencies of 20.43 cd A⁻¹ and 18.33 lm W⁻¹, respectively, and CIE coordinates of (0.62,

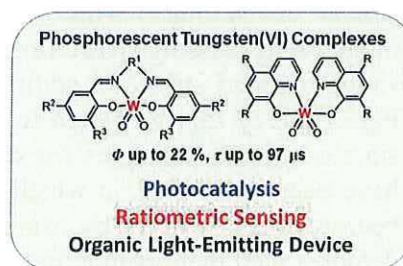


0.36) have been achieved. In addition, a significant reduction of efficiency roll-off is observed due to the bulky three-dimensional structure of the norborene moiety at the periphery of the Schiff-base ligand, maintaining high current efficiency of 14.69 cd A^{-1} and EQE of 8.3 % at the brightness of $1,000 \text{ cd m}^{-2}$. More importantly, *the fabricated red light-emitting device exhibits a long projected lifetime (T_{50}) of up to 18,000 hours*. This work has been published in *Chem. Asian J.* **2014**, *9*, 2984–2994.

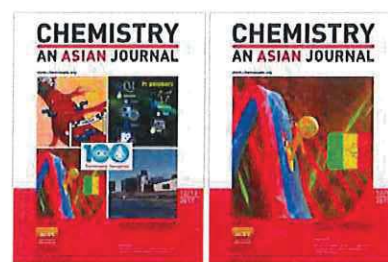
- By utilizing the modified device structure consisting of a composite blue host, both device efficiency and efficiency roll-off of platinum(II)-based white-emitting OLEDs (WOLEDs) can be significantly improved. The WOLED exhibits a two-fold enhancement in total power efficiency from 30 lm W^{-1} up to 61 lm W^{-1} and a reduced driving voltage at a luminance of $1,000 \text{ cd m}^{-2}$ from 10 V to 7.5 V. In addition, the efficiency roll-off at high brightness can be greatly reduced. Performance improvement can be rationalized in terms of a broadened exciton formation zone and prevention of exciton accumulation at the organic/organic contact for reduced triplet-triplet annihilation. *This finding provides a simple approach for improving the performance of WOLEDs without the use of complicated out-coupling techniques and has been published in Adv. Funct. Mater.* **2013**, *23*, 5168–5176 and highlighted as frontispiece.



- The development of strong luminescent metal complexes from inexpensive earth-abundant metals is attractive for low-cost solution-processable OLEDs, especially for large-area displays. With efforts dedicated by the team, *a new class of air-stable luminescent tungsten(VI) complexes with room temperature emission quantum yields up to 0.22 in thin films has been demonstrated for the first time*. Surprisingly, changing the supporting ligand allows modulation of intersystem crossing efficiency and dual fluorescence-phosphorescence with comparable intensities has been realized. Solution-processable OLEDs based on tungsten(VI) complexes show a stable yellow emission with EQEs of up to 4.8 %. It is conceivable that air-stable phosphorescent tungsten complexes could become as competitive as other luminescent complexes of iridium(III) and platinum(II). The absence of a deactivating d-d ligand field excited state in tungsten(VI) may provide an advantage over other metal complexes when materials with high-energy phosphorescence are desired. This finding has been published in *Angew. Chem. Int. Ed.* **2017**, *56*, 133–137.



- Phosphors based on another cheap and abundant metal have also been successfully designed and synthesized. Particularly, a series of 7,8-bis(diphenylphosphino)-7,8-dicarbano-undecaborate and neocuproine-based copper(I) complexes with tunable emission colors from green to deep red has been synthesized via rational modification of the neocuproine ligand structure. Photophysical and computational studies have been performed to elucidate the electronic structures and the transition features of the complexes, underlining the geometric and electronic factors of the ligand in determining the emission properties of the complexes. Notably, efficient solution-processable OLEDs with emission spanning from green to red and maximum EQEs of 18.5 %, 14.3 %, and 10.2 %, respectively, have been achieved. *These results have been published in Chem. Asian J.* **2017**, *12*, 1490–1498 in the special issue celebrating the 100th anniversary of the founding of Royal Australian and highlighted as Front Cover and Back Cover.

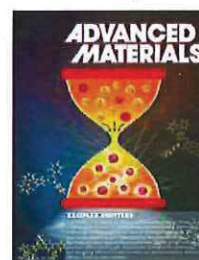


- Apart from the phosphorescent materials, the development of TADF materials has recently aroused enormous interest because of their capability to harvest all singlet and triplet excitons for the realization of 100 % internal quantum efficiency in OLEDs. In collaboration with Prof. Xiao-Hong Zhang from Technical Institute of Physics and Chemistry in Beijing, a simple approach for the realization of nearly 100 % triplet exciton harvesting in conventional fluorescent

dopant-based OLEDs has been developed. This approach has been successfully demonstrated by using a TADF exciplex host to up-convert the triplet excitons of fluorescent materials to the singlet states. **By choosing a suitable low dopant concentration to minimize the short-range triplet-triplet energy transfer from the host to the guest, the fraction of excitons that are capable of radiative decay can be significantly increased up to 83.1 %.** The best 2,3,6,7-tetrahydro-1,1,7,7-tetramethyl-1*H*,5*H*,11*H*,10-(2-benzothiazolyl)quinolizino-[9,9*a*,1*gh*]coumarin-doped device employing an exciplex host exhibits a maximum current efficiency of 44.0 cd A⁻¹, a maximum power efficiency of 46.1 lm W⁻¹, and an impressive EQE of 14.5 %. **This is the first OLED using a conventional fluorescent dopant sensitized with a TADF exciplex host.** These findings reveal that it is feasible to harvest triplet excitons in conventional fluorescent dopant-based OLEDs through energy transfer from exciplex host. **This finding has been published in *Adv. Mater.* 2015, 27, 2025–2023 and highlighted as Frontispiece.**



- Continuous work on the development of TADF exciplex emitters has been carried out, in which exciplex emitters have been designed by mixing commonly used hole-transporting arylamines with a bipolar host with a high triplet energy. Particularly, one of these exciplex emitters, i.e. TAPC:DPTPCz, shows a high fluorescent quantum yield of 0.68 and a small singlet-triplet energy splitting (ΔE_{ST}) of 47 meV. This undoubtedly allows TADF emission via reverse intersystem crossing (RISC) from non-radiative triplet excited states to radiative singlet excited states. In addition, **the TAPC:DPTPCz-based device exhibits a high EQE of 15.4 %, which is the highest EQE reported for exciplex OLEDs.** More importantly, simple rational strategies for designing exciplex emitters with high fluorescent quantum yields have been developed, in which an efficient exciplex system and its resulting photon energy have been readily predicted by using the solution redox potentials of the constituting molecules. It is demonstrated that organic materials with high triplet energies are preferred for designing exciplex emitters with high fluorescent quantum yields as well as enabling efficient RISC. **This work has been published in *Adv. Mater.* 2015, 27, 2378–2383 and highlighted as Frontispiece.**



- Through collaborative efforts, these TADF exciplex emitters have been utilized to generate white light emission. Efficient TADF emission can be realized by careful material selection; specifically, a novel efficient TADF blue exciplex is formed by mixing hole-transporting molecule 4,4'-bis(9-carbazolyl)-2,2'-dimethylbiphenyl (CDBP) with an electron-transporting molecule ((1,3,5-triazine-2,4,6-triyl)tri(benzene-3,1-diyl))tris(diphenylphosphine oxide) (PO-T2T) with a ratio of 1:1. The CDBP:PO-T2T blue exciplex shows efficient TADF emission and intrinsically high triplet energy, making itself an excellent candidate for both blue emitter and triplet host for green and red phosphors. Highly efficient blue-emitting devices based on this TADF exciplex emission and green- and red-emitting PHOLEDs with extraordinarily high EQEs of 13.0 %, 28.6 %, and 28.0 % have been successfully prepared. More importantly, a single emissive layer hybrid WOLED with a low turn-on voltage of 2.5 V as well as maximum forward-viewing current, power efficiencies, and EQEs of 67.0 cd A⁻¹, 84.1 lm W⁻¹, and 25.5 % using light out-coupling technique has also been realized. The excellent performance is ascribed to the “barrier-free” architecture and a bipolar emissive layer arising from the exciplex system. This work not only can offer a feasible approach for preparing high-performance single emissive layer hybrid WOLED for large-area and low-cost applications, but also can overcome the drawbacks on the low performance of the conventional single emissive layer hybrid WOLEDs based on blue fluorescent hosts. **This finding has been published in *Adv. Mater.* 2015, 27, 7079–7085 and highlighted as Frontispiece.**



2. Development of OPV Materials

Collaborative team efforts have generated different new classes of high-performance photoactive materials as well as smart device architecture for efficient organic photovoltaic (OPV) devices and perovskite solar cells. Especially, **the team has demonstrated the fabrication of ultrathin and**

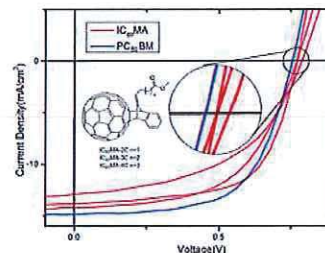
flexible perovskite solar cells with graphene transparent electrodes for the first time, the use of solution exfoliated few layers black phosphorous as an electron-transporting layer in OPV devices for the first time, and the first report on the synthesis of perovskite films by thermal annealing of evaporated lead(II) iodide/methylammonium iodide multilayers. The pioneering works of the team have also been invited to write review articles for the development of graphene and other 2D materials in *Chem. Soc. Rev.* and the rational electron-donating side chain engineering for OPV devices in *Macromol. Rapid Commun.* In addition, the team has explored various convenient and cost-effective techniques for realizing high-performance OPV devices as well as simple approaches for achieving pinhole-free perovskite films with high photostability. Some of the works have been highlighted as Front Cover pages and Frontispiece. ***The team has also developed low-cost high-purity separation processes for fullerene, in which the production cost of 1 gram of the fullerene with 99.93 % purity is much lower than those commercially available from the suppliers in the market.***

Below are listed the selected outputs related to the deliverables of the project:

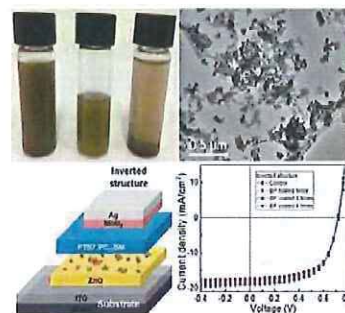
- While the use of molecular materials having long-lived triplet excited states for harvesting solar energy could be an effective approach to boost up the power conversion efficiency (PCE) of OPV devices, the performance of this kind of devices as reported in the literature is still rather low. Attempts have been made to explore metal-organic/organometallic complexes for the fabrication of efficient OPV devices. Three gold(III) corroles, **Au-C1**, **Au-C2** and **Au-C3**, have been designed and synthesized, in which these gold(III) corroles display relatively long triplet excited state lifetimes of $\geq 25 \mu\text{s}$ and low emission quantum yields of $< 0.15 \%$. By blending the gold(III) corroles as electron-donor with electron-accepting fullerene, the optimized vacuum-deposited OPV device based on **Au-C2** gives high short-circuit current density (J_{sc}) of 9.21 mA cm^{-2} , an open-circuit voltage (V_{oc}) of 0.82 V , and a fill factor (FF) of 0.40 , corresponding to a high PCE of 3.0% , under AM 1.5 sunlight illumination. The PCE can be further boosted up to 4.0% after thermal treatment of the OPV device. Solution-processed OPV device based on **Au-C2** with a high PCE of 6.0% has also been prepared. ***These PCE values are among the best reported for both types of vacuum-deposited and solution-processed OPV devices fabricated with metal-organic complexes having long-lived excited states as electron-donor. These findings have been published in Adv. Funct. Mater. 2014, 24, 4655–4665 and highlighted as Frontispiece.***
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- The team has demonstrated exciting works on the development of low bandgap polymers for light harvesting and ***has been invited to write a review article to summarize the rational electron-donating side chain engineering for improving the performance of OPV devices.*** A survey of representative examples which use electron-donating alkylthio and alkoxy side chains in conjugated organic polymers for polymer solar cell applications has been presented. It is found that engineering alkylthio and alkoxy side chains is an easy and effective strategy to fine-tune the absorption spectra and energy levels of the conjugated polymers, which should be kept in mind when constructing π -conjugated backbone of photovoltaic materials. The performance of OPV devices is highly dependent on the polymeric backbone planarity, molecular packing and film morphology, and hence modulating the side chain is a complex study. A better understanding of this kind of side chain behaviour in
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solution-processable conjugated organic polymers will greatly facilitate the development of the polymer donor materials. This work has been published in *Macromol. Rapid Commun.* **2016**, *37*, 287–302 and **highlighted as the Front Cover page**.

- PC₆₁BM and PC₇₁BM are the prototypical acceptor materials for solution-processed OPV devices because of their outstanding solubility and extremely high electron mobilities. The team has devoted the collaborative efforts on the development of new acceptors with high lowest unoccupied molecular orbital (LUMO) levels to replace PCBM. A new design of the fullerene derivatives, which combines the LUMO up-shifting property of indene-C₆₀-bisadduct (ICBM) with the improved processability via incorporation of alkyl side chain with an ester group from PCBM, has been employed to synthesize a series of isobenzofulvene mono-adduct fullerene derivatives (IC₆₀MA-2C, IC₆₀MA-3C and IC₆₀MA-4C). These compounds exhibit upshifted LUMO levels compared with PC₆₁BM and excellent solubility in most of the common solvents. These compounds work well with poly[4,8-bis[(2-ethylhexyl)-oxy]-benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]-thiophenediyl] (PTB7). All OPV devices based on these IC₆₀MA derivatives have shown an increased V_{OC} than those of the PC₆₀BM-based devices. Photovoltaic devices based on the blend of PTB7:IC₆₀MA-4C give a high PCE of up to 6.5 % whereas PTB7:IC₆₀MA-3C based OPV devices exhibit the highest V_{OC} value of 0.79 V among them. Electron mobilities of these fullerene derivatives have been measured, in which high electron mobilities of up to $10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, contributing to the high FF observed in the OPV devices. **These IC₆₀MA derivatives are attractive alternative electron acceptors to PC₆₁BM with up-shifted LUMO levels and decent material processability for device fabrication.** This finding has been published in *J. Mater. Chem. C* **2015**, *3*, 977–980.

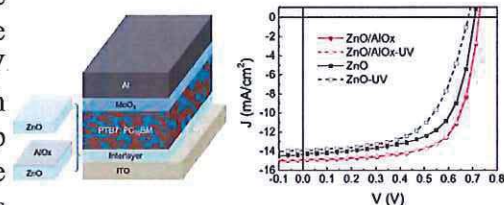


- In addition to the development of novel OPV materials, various smart device architectures have been explored in order to further improve the performance of OPV devices. Collaborative team efforts have led to the development of high-performance device architecture that can substantially enhance the efficiencies of OPV devices. While most of the 2D materials serve as hole-transporting layers in organic optoelectronic devices, **the team has demonstrated the use of solution exfoliated few layers black phosphorus (BP) as an electron-transporting layer in OPV devices for the first time.** Specifically, solution exfoliated few layers BP are 2D van der



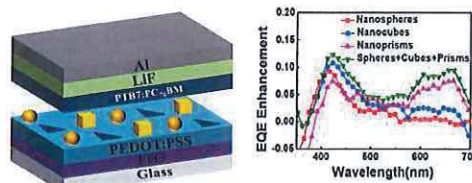
Waals crystals that possess excellent electronic and physical properties. It is found that the incorporation of BP flakes with optimum thickness of $\sim 10 \text{ nm}$ can form cascaded band structure in OPV devices, which can facilitate electron transport and enhance the PCEs of the devices. Particularly, the PCEs of the BP-incorporated OPV devices can be improved to 8.2 % in average with the relative enhancement of 11 %. In addition, the BP incorporation can improve the stability in air due to the encapsulation of the devices and are thus suitable for practical applications in the future. These findings have been published in *Adv. Funct. Mater.* **2016**, *26*, 864–871.

- Through the collaborative team efforts, we have developed a new approach to improve the charge extraction and performance reproducibility of the OPV devices via the modification of the zinc oxide (ZnO) with an ultrathin aluminium layer for suppressing the subgap states, where their presence will dramatically hamper the charge collection efficiency. As confirmed by various characterization techniques, including photothermal deflection spectroscopy, high-resolution X-ray photoelectron spectroscopy and dark-current measurements, the suppression of ZnO subgap states by modification of its surface with an ultrathin Al layer can significantly improve the charge extraction, achieving PCE of 8.0 % for the PTB7:PC₇₁BM-based inverted OPV devices, which is

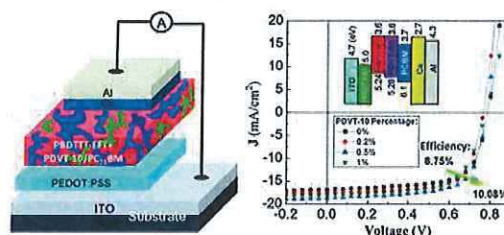


~15 % higher than that of a structurally identical control cell made with a pristine ZnO interlayer. More importantly, *such Al-modified OPV devices exhibit high-performance reproducibility, in which their photovoltaic responses are almost constant under different surface treatment conditions, e.g. with intense UV exposure.* These findings verify that the use of Al-modified ZnO interlayer favors the efficient charge collection, and provide a low-cost and simple solution-based preparation process for producing high-performance OPV devices. This work has been published in *ACS Appl. Mater. Interfaces* **2016**, *8*, 14717–14724.

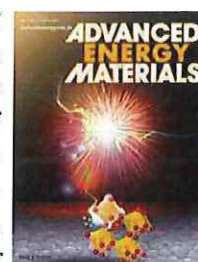
- Another simple and new approach for attaining light absorption enhancement in OPV devices via the introduction of silver nanoparticles (Ag NPs) with different shapes of nanosphere, nanocube and nanoprism has been demonstrated. It is found that the OPV devices having a combination of different shaped Ag NPs are more favorable for efficient light harvesting. The use of the mixed Ag NPs ethanol solution benefits the performance of the OPV devices by enhancing light absorption in the PTB7:PC₇₁BM active layer through the coupling of localized surface plasmon resonance and by facilitating homogeneous vertical phase separation favoring the exciton dissociation, charge transport and collection. *The EQE spectrum of the OPV devices incorporated with the tri-shaped Ag NPs can be significantly enhanced at a very broad wavelength range of 370–700 nm and the enhancement amplitude is much higher than that with any single-shaped Ag NPs.* The Ag NP-doped device exhibits high PCE of 8.0 %, much higher than that of the optimized control cell (6.7 %) made without Ag NPs. This finding has been published in *Mater. Today Energ.* **2017**, *3*, 84–91.



- Collaborative team efforts have further led to a pronounced enhancement in PCE of OPV devices. By adding a suitable high-mobility polymer of only 0.5 wt%, an obvious improvement of EQEs over a broad wavelength range from 350 nm to 800 nm can be observed. The PCE of the OPV devices based on poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)-benzo[1,2-*b* ;4,5-*b'*]dithiophene-2,6-diyl-*alt*-(4-(2-ethylhexyl)-3-fluorothieno[3,4-*b*]thiophene)-2-carboxylate-2,6-diyl] and PC₇₁BM can also be increased from 8.75 % to 10.08 %. The drastic enhancement in PCE can be attributed to an increased hole mobility and carrier lifetime in the devices, as evidenced by the impedance studies. It is noticed that the additive should have a similar highest occupied molecular orbital (HOMO) level to that of the donor material to form a cascade band structure; particularly, several tens of millivolts on HOMO level may lead to a completely different effect on the photovoltaic responses. *Taking the advantages of a plenty of choices of high-mobility polymers on the market, this work provides a convenient and cost-effective technique for realizing high-performance OPV devices. These findings have been published in Energy Environ. Sci.* **2015**, *8*, 1463–1470.



- Among all emerging solar cell technologies, the development of perovskite solar cells has been a red-hot topic in both academic and industrial research due to their exceptionally promising performance surpassing all alternative technologies and approaching to the inorganic silicon counterparts. While record-high PCEs of up to 22.1 % have been certified for perovskite solar cells, poor reproducibility and lack of long-term stability of these hybrid solar cells have to be overcome before the cells can be commercialized. The team has performed a deep investigation on the stability of perovskite solar cells. *Unlike the common belief that excess lead iodide (PbI₂) can be beneficial to the efficiency of perovskite solar cells, it is found that a small amount of unreacted PbI₂ results in an intrinsic instability of the perovskite film under illumination, leading to the film degradation under inert atmosphere and faster degradation upon exposure to illumination and humidity.* The perovskite films without PbI₂



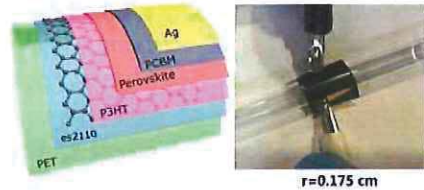
have improved stability, but lower efficiency due to inferior film morphology in terms of smaller grain size and the presence of pinholes. Optimization of the deposition process for PbI₂-free films leads to dense, pinhole-free, large grain size perovskite films which result in cells with high efficiency without detrimental effects on the film photostability. *This work has been published in Adv. Energ. Mater. 2016, 6, 1502206 and highlighted as Front Cover page.*

- While 2D graphene and its oxide have been demonstrated to be ideal materials as transparent electrodes or interlayers in almost all types of photovoltaic devices, great challenges still remain unsolved and are the biggest obstacles to commercialization.

With the efforts dedicated by the team, *we have demonstrated the fabrication of ultrathin and flexible perovskite solar cells with graphene transparent electrodes for the first time.* The flexible devices with the structure of polyethylene terephthalate/graphene/P3HT/methylammonium lead iodide

(CH₃NH₃PbI₃)/PC₇₁BM/Ag are prepared on 20 μm-thick polyethylene terephthalate substrates by low-temperature solution process, which show the PCE of 11.5 % and high bending durability.

Moreover, the devices demonstrate the power output per weight of ~5 W g⁻¹, which is much higher than those of conventional inorganic solar cells (0.1–0.4 W g⁻¹). This work paves a way for preparing flexible perovskite solar cells as well as other optoelectronic devices by using graphene transparent electrodes. These findings have been published in *Nano Energ. 2016, 28, 151–157.*



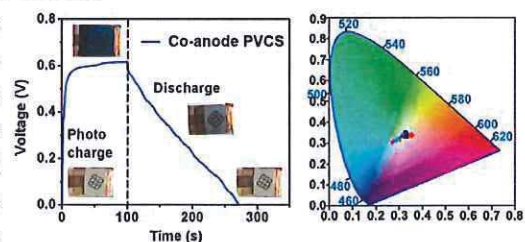
- A comprehensive review on the recent advances in the applications of graphene and other 2D materials in various types of solar cells, including OPV devices, Schottky junction solar cells, dye-sensitized solar cells, quantum dot-sensitized solar cells, perovskite solar cells, and other inorganic solar cells, mainly focusing on the critical issues of device design and processing techniques in using these materials has been performed. For practical applications, the device performance should be improved by optimizing the 2D material synthesis, film transfer, surface functionalization and chemical/physical doping processes, especially for the fabrication of large-area devices. *This result summarizes the recent progress of graphene and other 2D materials and offers guiding principles for the development of these materials for high-performance solar cells and has been published in Chem. Soc. Rev. 2015, 44, 5638–5679.*



- The team has also been invited to write *a review article on the developments in the field of semi-transparent solar cells with a view to inspire more fantastic ideas on material synthesis and device design that will lead to major progress in the fabrication and application of these emerging semi-transparent solar cells.* Particularly, this review article summarizes the advances in the preparation of semi-transparent solar cells, dye-sensitized solar cells, and polymer solar cells, focusing on the top transparent electrode materials and device designs, which are all crucial to the performance of these devices. Techniques for optimizing the efficiency, color and transparency of the devices are addressed in detail. Finally, a summary of the research field and an outlook into the future development in this area are provided. Due to their unique properties, these emerging semi-transparent solar cells can provide not only efficient power-generation but also appealing images and show promising applications in building integrated photovoltaics, wearable electronics, photovoltaic vehicles and so forth in the future. This work has been published in *Adv. Mater. 2017, 29, 1700192.*

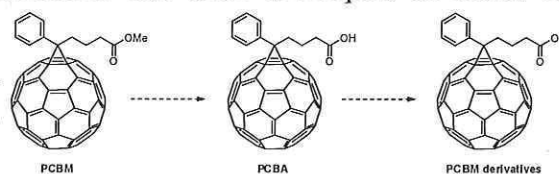


- In order to demonstrate the superior performance of the perovskite solar cells, attempts have been made to explore the integration of energy harvesting and storage devices in the construction of novel optoelectronic devices. Specifically, the team has demonstrated co-anode and co-cathode photo-voltachromic super-capacitors (PVCSs) by vertically integrating a perovskite solar cell with molybdenum oxide (MoO₃)/Au/MoO₃ transparent electrode and



electrochromic supercapacitor. *The PVCSSs provide a seamless integration of energy harvesting/storage device, automatic and wide color tunability, and enhanced photostability of perovskite solar cells.* Compared with conventional photovoltachromic cells, the counter electrodes of our PVCSSs provide sufficient balancing charge, eliminate the necessity of reverse bias voltage for bleaching the device, and realize reasonable *in situ* energy storage. The color states of PVCSSs indicate the amount of energy stored and energy consumed in real time, and enhance the photostability of photovoltaic component by preventing its long-time photo-exposure under fully charged state of PVCSSs. This work designs PVCSS devices for multifunctional smart window applications commonly made of glass and has been published in *ACS Nano* **2016**, *10*, 5900–5908.

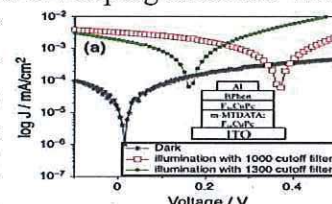
- The team has also devoted the efforts on the development of low-cost high-purity separation processes for fullerene derivatives. Post-modification of PCBM via saponification or esterification that could provide a rapid access to PCBM derivatives has been developed. A series of fullerene-containing polymers has been designed and synthesized by incorporating vinyl groups onto the fullerene derivatives through hydrolysis of PCBM in acid solution. Through the reduction of PCBM with hydroxy group, another fullerene derivative can also be produced with high yield of up to 72 %. Notably, the production cost of 1 gram of the fullerene with 99.93 % purity is much lower than those commercially available from the suppliers in the market.



3. Interfacial and Dynamic Studies

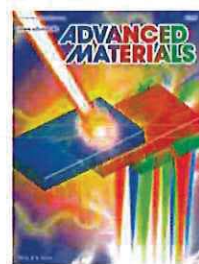
While interfacial energetics and charge interactions at the organic heterojunctions play important roles influencing device performance and lifetimes of the OLEDs and OPVs, current knowledge on their photocharge generation mechanisms, especially at the perovskite/fullerene junction, is very limited. *The team has demonstrated for the first time a simple approach for harvesting near-infrared photons with wide-energy-gap materials by making use of their interactive charge-transfer complex (CTC).* This finding generates new knowledge on the charge transfer interface and have important implications for boosting up the OPV device performance and stability. *A new approach for fabricating a wide-spectral photoresponse photo-detector via the preparation of black-color molybdenum oxide (BMO) has also been demonstrated for the first time,* which endow the originally wide-bandgap material with optical properties suitable for applications in the UV-visible-IR range. Meanwhile, the team has provided important information on the formation chemistry of perovskite/fullerene junctions as well as the controlling factors governing the photovoltaic responses and degradation mechanisms in OPV devices. Below are listed the selected outputs related to the deliverables of the project:

- Exploitation of energy from the NIR region is one strategic approach for enhancing the performance of OPV devices. While effort has been mostly put toward developing materials with narrow energy gaps, *our team has developed a simple approach for harvesting NIR photons with wide-energy-gap materials by making use of their interactive CTC.* Through photoemission studies, the interface between MoO₃ and rubrene was found to possess an abrupt discontinuity in the vacuum level, resulting in significantly overlapped electron wavefunctions and CTC formation. The CTC induces an intermediate state within the original energy gap of rubrene with energy of ≈ 1.3 eV, suggesting the feasibility of a charge transfer exciton generated upon NIR excitation. These results have been substantiated via device fabrication studies. This is confirmed by the generation of electric power OPV devices with an active layer of MoO₃:rubrene composite under excitation with a NIR light source. This finding has been published in *Adv. Funct. Mater.* **2012**, *22*, 3035–3042. This study opens up new research windows for NIR charge generation based on CTC formed between



wide-energy-gap materials. ***It is the first report of an OPV device which harvests IR solar energy via absorption of CTC formed by two small molecular materials.***

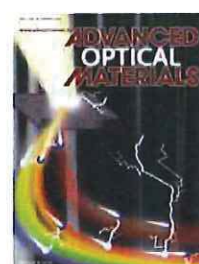
- Extension of this pioneering work has been carried out to further explore the feasibility of exploiting the NIR region of the solar spectrum for electric power generation using wide-energy-gap materials. Particularly, ***the team has been invited to write a Review Article to highlight the recent progress on the surface charge interactions and electronic structures of CTCs***, as well as to review the role on exciplex emission and potential applications for the generation of NIR photocharges using wide-energy-gap semiconductors. It has been demonstrated that CTCs can be used for NIR photovoltaic devices via sub-energy-gap electronic transitions using wide-energy-gap semiconductors with strong charge coupling. This can provide new insights for exploiting charge-interactive couples for new device applications, as well as can lead to breakthroughs regarding the understanding of the physical processes in CTCs and rational design concepts of CTC-enabled optoelectronic devices. ***This work has been published in Adv. Mater. 2014, 26, 5569–5574 and highlighted as Inside Front Cover.***



- Poor operation stability is a major hurdle for the wide application of OPV devices. While most attention is given to environmental threats to device stability, the team has attempted to investigate the intrinsic factors contributing to the degradation by X-ray photoemission spectroscopy (XPS) analysis. Although impressive device performance has been achieved for boron subphthalocyanine chloride (SubPc)/fullerene based device, the forming boride bond at its interface hinders the interfacial exciton dissociation and leads to device deterioration. Due to the high electron affinity of MoO₃ film, the incorporation of MoO₃ layer under the SubPc film has strong electron-drawing property and leads to CTC formation at the MoO₃/SubPc interface. The resulting charge redistribution in SubPc molecules effectively suppresses the further interfacial reaction at the SubPc/C₆₀ junction. Particularly, a significant degradation on EQE over the entire visible range has been observed after 1-hour irradiation. In sharp contrast, the MoO₃/SubPc/C₆₀ device shows limited (2 %) and negligible (0 %) drop in main signal peaks of both C₆₀ and SubPc, respectively. ***This work is the first report on the time-dependent interfacial degradation of OPV devices and the first demonstration that an interfacial chemical reaction donor/acceptor junction can be controlled by charge modulation and carrier redistribution. These findings have been published in Adv. Mater. Interfaces 2014, 1, 1300082 and highlighted as Inside Front Cover page.***

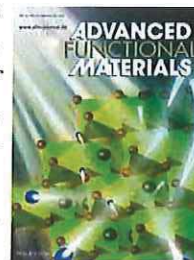


- A new approach for fabricating a wide-spectral photoresponse photo-detector via the preparation of black-color molybdenum oxide (BMO) has been demonstrated. With a simple annealing process in vacuum, a substantial color change from transparent to black is observed in the pristine MoO₃ film. Evidence from UPS and XPS studies illustrates the emergence of additional energy levels in the BMO with a long tail extending from its valence band. These forming gap states effectively split the bandgap of MoO₃ into two regions of approximately 1.8 and 1.0 eV, for two successively photon harvesting. By putting hole- and electron-extracting organic materials with matched energy levels aside to BMO, a photo-detector exploiting both the 1.8 and 1.0 eV sub-bandgap photon absorptions has been demonstrated. The device exhibits a wide spectral response from 320 to >1000 nm, manifesting applications directly inherited from the novel “black-color-oxide”. ***These findings demonstrate for the first time the black color of BMO can indeed endow the originally wide-bandgap material with optical properties suitable for applications in the UV-visible-IR range. This work has been published in Adv. Optical Mater. 2013, 1, 699–702 and highlighted as Back Cover Page.***

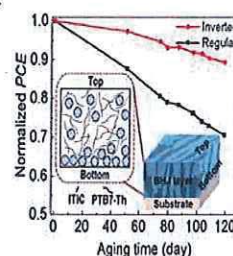
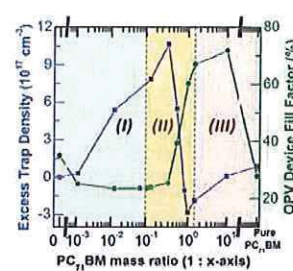
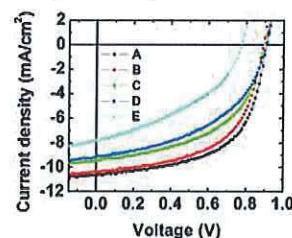


- The team has devoted its efforts to investigate the interfacial electronic structures of vacuum-deposited perovskite/fullerene and solution-processed perovskite/PCBM junctions by photoemission spectroscopies. The perovskite/fullerene junction behaves as a distinct N–N junction instead of a presumed P–N junction. Such type I N–N junction with straddling gap is

electronically inert and carries no charge interactions in the vicinity of the forming junction. *These findings provide a clear evidence that free carriers can be generated within the perovskite absorber with a very small binding energy of ~50 meV*, where the photogenerated excitons in the perovskite materials can be dissociated upon photoexcitation. Steady-state photoluminescence spectroscopy and OPV device fabrication have been performed to further verify the occurrence of an instant charge transfer at perovskite/fullerene junctions without any assistance from a P–N junction. *This work has been published in Adv. Funct. Mater. 2015, 25, 1213–1218 and highlighted as Frontispiece.*



- Batch-to-batch variations usually occur in the commercially available organic OPV materials and this inevitably results in large fluctuations in device performance from laboratory to laboratory. This batch-to-batch variation is a key obstacle for the commercialization of this emerging technology in the future. Through collaborative efforts, the team has carried out a detailed investigation of the impact of molecular weight distribution of a well-known photoactive donor material, poly[*N*-9'-heptadecanyl-2,7-carbazole-*alt*-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)] on photovoltaic device performance and carrier transport properties. It is found that different batches of polymers have substantial differences in their molecular weight distribution. Photovoltaic devices fabricated with a higher proportion of low molecular weight component have PCEs reduced from 5.7 % to 2.5 %. The corresponding charge carrier mobility at the short-circuit region is also significantly reduced from 2.7×10^{-5} to 1.6×10^{-8} $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$. These results have shown that an additional small molecular weight component of the polymer would significantly reduce both the device efficiency and carrier mobility. The reduction of mobility is due to the larger hopping distances arising from the additional small molecular weight component, as analysed by the Gaussian disorder model. *This work not only provides a detailed analysis of the effect of batch-to-batch variation of photoactive materials on device performance, but also provides a physical origin of the difference in carrier mobility and the resulting photovoltaic device performance.* This work has been published in *Adv. Energy Mater.* **2014**, 4, 1400768.
- In collaboration with Prof. Franky So from North Carolina State University, the team has examined the key limiting factors that control the device performance of OPVs in unoptimized regime, where the tuning of donor-acceptor weight ratio is an essential step to maximize the photovoltaic responses of bulk heterojunction solar cells. It is found that the creation of traps and localized states originated from fullerene molecules is the key limiting factor. With the aid of photothermal deflection spectroscopy, the trap density in the bulk heterojunction can be effectively quantified. Addition of small concentration of fullerene increases the electron trap density and lowers the electron mobility. When the donor-acceptor weight ratio reaches 1:0.1, fullerene percolation occurs. *There is an abrupt drop in trap density and simultaneously six orders of magnitude increases in the electron mobility.* Furthermore, the *FF* of the corresponding OPV devices are found to anti-correlate with the trap density. This study reveals that electron trapping is the key limiting factor for unoptimized bulk heterojunction solar cells in low fullerene regime. This work has been published in *Adv. Energ. Mater.* **2017**, 7, 1602360.
- To date, high performing nonfullerene OPV devices with inverted architecture are typically adopted; however, the advantages of suppression of bimolecular recombination and enhancement of charge extraction, which underpin the optimal cell performance and operational stability of the inverted nonfullerene OPV devices, have not yet been studied. The team has attempted to unravel the origin of efficient operation of nonfullerene OPV devices by using a combination of the transient photocurrent and light intensity-dependent current density–voltage characteristics, as well as XPS analysis to study the interfacial properties.



It is found that high performing inverted OPV devices benefit from the combined effects of suppression of bimolecular recombination enabled by an augmented effective internal electric field and improvement of charge extraction by avoiding the holes passing through the acceptor region, which would otherwise occur in a regular configuration cell. *The inverted OPV devices possess a significant improvement in the cell stability and a high PCE of 8.0 %, which is >29 % higher than that of an optimized regular configuration control cell (6.1 %).* The combined effects also lead to a slow cell degradation process. This finding has been published in *J. Phys. Chem. Lett.* **2017**, 8, 5264–5271.

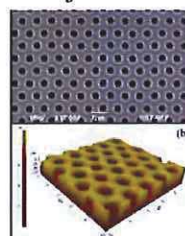
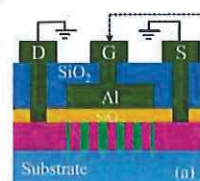
4. Development of Industrial-Competitive Technologies for OLEDs and OPVs

Collaborative team efforts have led to the development of industrial-competitive technologies for OLEDs and OPVs as well as the realization of highly efficient large-area OLEDs and OPVs meeting the promising deliverables. Notably, *the team has developed a pioneering bridged-grain (BG) technique that can be applied in the fabrication of low-temperature polycrystalline-silicon (LTPS) thin film transistors (TFTs), i.e. an essential element for large-area active-matrix OLEDs (AMOLEDs). The team also demonstrates for the first time the use of submicron-dot-array doped channel for the fabrication of solid-phase-crystallized poly-Si TFTs.*

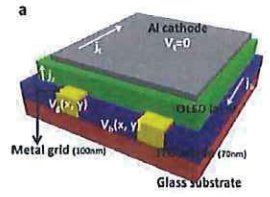
These breakthrough technologies can significantly improve the current flow, and the fabricated TFTs exhibit low threshold voltage, high field-effect mobility, low leakage current and larger on/off ratio.

On the other hand, *the team has successfully made use of the simple and well-established closed space vapor transport (CVST) technology for the fabrication of highly efficient perovskite solar cells on large-area substrates*, providing a reliable and scale method paving the way for the commercialization of perovskite solar cells. Below are listed the selected outputs related to the deliverables of the project:

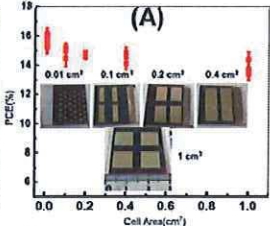
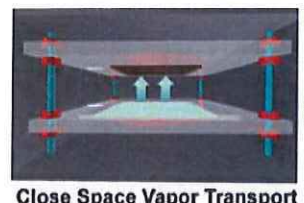
- Collaborative team efforts have led to the pioneering development of a BG TFT structure. Specifically, the BG structure can enhance the electrical characteristics in terms of sub-threshold slope and threshold voltage of the LTPS TFTs, as well as provides a simple and low-cost method for the realization of large-area OLEDs. The team has further devoted the efforts to investigate the device degradation of BG polycrystalline silicon (poly-Si) TFTs for the first time. It is found that the BG TFTs are much more stable than normal TFTs under the same dynamic negative bias stress (NBS); particularly, the dynamic NBS-induced hot carrier degradation is significantly reduced from -99.9% to -2.4% after 10^4 dynamic NBS. The sharing of the electric field across multiple reverse biased junctions is the major reason responsible for the improved dynamic hot-carrier reliability in the BG TFTs. *These results indicate that this BG structure can suppress the hot carrier effect, which has great potential for system-on-panel application in the future.*
- In order to realize large-area devices, extension of the work has been carried out on the development of a novel “submicron-dot-array” doping method for the fabrication of LTPS TFTs, in which the submicron-dot-array structure was formed by laser interference lithography. All key TFT parameters, including sub-threshold slope, on-state current/off-state current ratio, have been greatly improved. A high field-effect mobility of up to $58.4\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ can be obtained for poly-Si TFTs with submicron-dot-array structure, which is four times larger than that of conventional poly-Si TFTs. Furthermore, the method is low temperature compatible, two dimensional and without process variation and reliability issues, and has great potential for system-on-panel applications. *This work demonstrates for the first time the use of submicron-dot-array doped channel for the fabrication of solid-phase-crystallized poly-Si TFTs.*



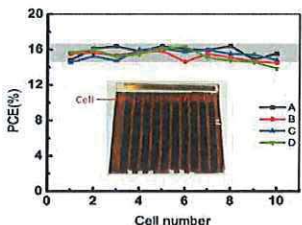
- Luminescence and temperature non-uniformity are serious problems as the potential drop across the large indium-tin-oxide (ITO) electrodes becomes large. Moreover, larger emitting area is more sensitive to the defects introduced during the fabrication process, which cause the current leakage and even the detrimental short circuit. These serious problems lead to the challenges for the realization of large-area OLEDs in the commercial products. The team has carried out a comprehensive study on the geometrical dimensional relationship of the metal grid on a large-area OLED light panel electrode. A method to find the optimized solution of height and width under given operational voltages has been discussed, using differential equations and finite element method simulations. Experiments have been further performed to verify the simulation results. It is found by simulation that the optimization of height and width parameters can maximally reduce the inhomogeneity of OLED lighting panel. These findings provide an effective approach for patterning metal grid with low sheet resistance on the ITO electrode to get the best luminance uniformity for large-area OLED lighting panels.



- With the efforts dedicated by the team, a low-cost, high throughput and centimetre-scale fabrication method of efficient hybrid perovskite solar cells has been developed by closed space vapor transport (CSVT). CSVT is a well-established industrial technology for cost-competitive, commercial-scale manufacturing of polycrystalline CdTe solar cells. With the aid of CSVT process, we are capable to make efficient perovskite solar cells with PCEs of up to 16.2 %. Notably, *large-area devices with PCEs of 13.8 % and good uniformity in a large substrate of 3 cm × 3 cm have also been prepared.* This technique takes advantages of the relatively inexpensive and compact design, high throughput, efficient precursor transport/utilization and good large-area uniformity, indicating the potential of the CSVT process for manufacturing low cost, high throughput and large-area efficient perovskite solar cells in the future. This finding has been published in *Phys. Status Solidi RRL* **2016**, *10*, 153–157.



- Another simple and versatile strategy for the realization of high-efficiency and large-area perovskite solar cells has also been demonstrated. While nickel oxide (NiO_x) is widely used as surface modification layer for perovskite solar cells that can greatly improve the PCE of perovskite solar cells up to 19.2 %, its high-temperature process makes manufacturing more complex and hampers the development of perovskite-based tandem photovoltaics. The team has tackled the problem by using the magnetron sputtering, which is well established in the industry. The limitations of low *FF* and *J_{SC}* commonly observed in sputtered NiO_x-derived perovskite solar cells can be overcome through magnesium doping and low oxygen partial pressure deposition. *The fabricated perovskite solar cells show high PCEs of up to 18.5 %, along with negligible hysteresis, improved ambient stability, and high reproducibility. In addition, good uniformity is demonstrated over an area of 100 cm².* This simple and well-established approach constitutes a reliable and scale method paving the way for the commercialization of perovskite solar cells. This work has been published in *Adv. Sci.*, DOI: 10.1002/advs.201700463.



5. Granted Patents and Mid- to Down-stream R&D Activities

Through the efforts dedicated by the team, unique and benchmark contributions on the development of highly efficient OLED materials as well as patented technologies based on BG structure and low-cost, high-purity separation processes for fullerene derivatives have been successfully achieved and have generated Hong Kong-owned intellectual property (IP) rights and patents. *A total of 26 patents have been applied to US and other national patent officers, including China, Korea, and Europe; notably, 14 patents have been successfully granted.* These Hong Kong-owned IP rights, patents and technological know-how can definitely lead to the development of high technology R&D consumer electronics industries related to OLED and OPVs in Hong Kong, the Mainland and

the Guangdong Province region. The team has established a "HKU-TCL Joint Laboratory for New Printable OLED Materials and Technology" for collaboration and further development of our proprietary OLED materials.

6.2 What was the added value of the TRS funding, rather than standard project grant funding?

This TRS funding provides a great opportunity to foster close collaboration amongst chemists, physicists, materials scientists and engineers as well as world-leading international and national collaborations to meet the grand challenges related to OLEDs and OPVs and spanning from basic research to applied research activities, for which existing standard project grant funding cannot cater. This TRS funding has linked together the strengths in the various areas, various expertise, and various institutions and industries, locally, nationally and internationally, as reflected by more than 150 joint publications. In addition, the 5-year duration of this TRS funding enables the incubation and execution of research ideas and plans in material development which usually demands labour and time costs as fruitful research outcomes are often triggered by unexpected findings through continuous trial and proof. This TRS funding also provides a very good chance of delivering patentable materials or products of practical applications and technology transfer. Working closely with industries, more midstream and downstream research activities can be established for the realization of commercial products in the future.

6.3 If the project has not met its original objectives, why? N/A

6.4 (a) Peer-reviewed journal publication(s) arising directly from this project:

A total of 253 publications have been published for publications in international SCI journals, including *Energy Environ. Sci.*, *Mater. Today*, *Adv. Mater.*, *Adv. Energy Mater.*, *J. Am. Chem. Soc.*, *ACS Nano*, *Adv. Funct. Mater.* and *Angew. Chem. Int. Ed.* Among these publications, 89 publications are with SCI Impact Factor (IF) > 7.5, 44 publications with 7.5 > IF > 5, 40 publications with 5 > IF > 3.5. Of which 1 publication has been published in *Nano Lett.* as **Hot Paper** and **Highly Cited Paper** on ISI Web of Science. Among which 8 of them have been highlighted as Front Cover, 4 as Inside Front Cover, 4 as Back Cover, and 8 as Frontispiece, demonstrating the significance of these works. Over 150 joint publications have been published involving interdisciplinary or multi-institutional collaborations as a result of the collaborative efforts amongst team members and with local and national collaborators. Please refer to Appendix I for full list of publications and Appendix II for the highlights.

(b) Recognised international conference(s) in which paper(s) related to this project was/were delivered:

A total of 89 Invited Lectures related to the TRS project were presented in international/national conferences, among which 17 of these lectures were Plenary/Keynote/Award Lectures. Please refer to Appendix III for full list of invited lectures.

(c) RGC funding should have been acknowledged in all publication(s)/conference papers listed in (a) and (b) above. If no acknowledgement has been made in any of the publications/papers, please indicate and provide explanations. N/A

6.5 To what extent this project has strengthened inter-institutional collaborations and other partnerships?

Collaborative efforts amongst team members and international/national scientists have generated more than 150 joint publications involving inter-disciplinary and/or cross-institutional efforts. Selected highlights of the collaborative activities are listed as follows. V.W.-W. Yam (HKU) had collaborated with S.-K. So (HKBU) on the mobility measurements of spirothioxanthene compounds for the fabrication of OPVs. V.W.-W. Yam had also collaborated with C.-S. Lee (CityU) to develop a novel host material with carbazole-fluorenyl hybrid for orange-emitting OLEDs and warm WOLEDs as well as with H.-S. Kwok (HKUST) on possible realization of large-area OLEDs. C.-M.

Che (HKU) had collaborated with C. Surya (HKPolyU) on possible realization of large-area solar cells. H.-S. Kwok had been working in collaboration with M. Wong (HKUST) to develop novel BG technique and new approaches for LTPS TFTs as well as the investigation of degradation mechanisms. H.-S. Kwok had also collaborated with B.-Z. Tang (HKUST) on the development of an interesting class of aggregation-induced emission luminogens by tuning the singlet-triplet energy gap and with A. Rogach (CityU) to realize polarized emission from ordered semiconductor nanorods by the combination of photoinduced alignment and self-assembly. C.-S. Lee had collaborated closely with F.-R. Zhu (HKBU) to develop high-performance OLEDs and WOLEDs based on the TADF exciplex emission as well as a new approach for improving the performance of OPVs based on nanostructured pattern. C.-S. Lee had been working with Roy V.A.L. (CityU) on the development of controllable ambipolar flash memories and polymeric nanotubes with tunable aspect ratios and charge transport properties and with S.-K. So to achieve efficient OPVs by suppression of subgap states. Collaborative efforts among C.-S. Lee, C. Surya and F. Yan (HKPolyU) had led to the development of novel solution-processable ultrathin black phosphorus that is capable to serve as effective electron-transporting layer in OPVs. C. Surya had collaborated with W.-K. Chan (HKU) on the characterization of hydrothermally synthesized copper oxide as well as the development of efficient OPVs and planar perovskite solar cells. C. Surya had been working in collaboration with F. Yan to develop a new and simple approach for achieving highly efficient OPVs by adding a high mobility conjugated polymer and with S.-K. So on the crystal engineering and controlled deposition conditions for the realization of high efficiency hybrid perovskite solar cells. K.-M. Ng (HKUST) had been working in collaboration with S.-K. So on the fabrication of OPVs based on the newly developed isobenzofulvene-fullerene monoadducts and on the characterization of their electron mobilities. F.-R. Zhu had been working in collaboration with M.-S. Wong (HKBU) to develop a new class of heteroatom-substituted copolymers with bifunctional OPV and OTFT properties. W.-Y. Wong (HKBU) had collaborated with F. Yan on the synthesis of a new class of acceptor-donor-acceptor based small molecules for OPV applications. G.-H. Chen (HKU) had been working with the experimentalists to perform computational studies on the properties of their materials.

6.6 Research students trained (registration/awards):

Since the commencement of the TRS project in 2012, a total of **59** RPgs have been recruited. Amongst these students, **44** of them had graduated from their PhD studies and **3** of them had graduated from their MPhil studies. Details of these students are listed in Appendix IV.

6.7 Specific products (e.g. software or netware, instruments or equipment developed):

Since the commencement of the TRS project, a total of **26** patent applications have been filed, of which **14 patents have been successfully granted in US and national patent offices**. Please refer to Appendix V for the details of the filed and granted patents.

6.8 Other education activities and/or training programmes developed:

Education of students and the public in the science and technology of OLEDs and OPVs is one of the missions of the TRS project. A total of **59** RPg students have been registered and **47** of them had been graduated from their PhD/MPhil studies. In addition, three research symposia open to public had been held on 17–18 December 2013, 13 November 2014, and 8 January 2016 to bring in top scientists, including Prof. C.W. Tang from University of Rochester, Prof. Y. Yang and Dr. G. Li both from University of California, Los Angeles, Prof. C. Adachi from Kyushu University, Prof. Y.G. Ma from Jilin University, Prof. Y. Cao, Prof. H.B. Wu, Prof. F. Huang and Prof. H.L. Yip, all from South China University of Technology, to provide a forum for the exchange of ideas amongst top scientists in the fields of OLEDs and OPVs and team members. To facilitate team discussions and exchanges of research findings, four workshops involving all Co-PIs, Co-Is, collaborators and their RPg students and research personnel had also been held on 14 September 2013, 28 June 2014, 6 June 2015, and 16 August 2016 to ensure broad dissemination of results within the team. The poster announcements and scientific programme for the research symposia and workshops for team discussions and exchanges are included in Appendices VI and VII, respectively. A distinguished

lecture on “Evolution of the Next-Generation Display Technology” by Prof. C.-W. Tang from University of Rochester had also been organized to discuss the development pathway of OLED from early discovery to recent commercialization. V.W.-W. Yam had delivered an invited presentation at EmTech Hong Kong, i.e. MIT Technology Review’s Conference on Emerging Technologies that Matter, for the development of OLED materials in HKU on 6–7 June 2017 and a public lecture at the “Advances in Display Workshop”, co-organized by ASTRI and HKSTP, in December 2016 at Science Park. V.W.-W. Yam had collaborated with A. Rogach in organizing an international workshop on “Photonics of Functional Nanomaterials” that was held on 6–9 May 2013 at CityU to provide a forum for extensive exchange of ideas on the development of functional nanomaterials. C.-M. Che had collaborated with C.-S. Lee in organizing a conference on “International Conference on Molecular Electronic Materials and Devices (MEMD2015)” that was held on 5–8 January 2015 at CityU to bring in top scientists, including Prof. S.-R. Forrest (University of Michigan) and Prof. C. Adachi (Kyushu University), to provide a forum for extensive exchange of ideas on the development of molecular and polymeric optoelectronics. A. Rogach had also collaborated with the Nanosystems Initiative Munich (NIM) to jointly organize a “CFP-NIM Workshop on Functional Photonics and Nanosystems” at CityU on 27–29 May 2015 for a forum on the development of photonic materials on the nanoscale. Poster announcements and scientific programmes of distinguished lecture, “Photonics of Functional Nanomaterials Workshop”, “MEMD2015”, “CFP-NIM Workshop” have been listed in Appendices VIII–XI, respectively.

- 6.9 Please highlight any deliverables indicated in the project implementation timetable endorsed by the RGC which have not been covered or achieved as per sections 6.1 to 6.8 above, and explain/ elaborate. N/A

Project Management

6.10 Please elaborate how the PC has played his/her role in coordinating and managing the project. V.W.-W. Yam, the PC of the TRS project had performed her functions within the authority granted by the External Review Committee (ERC) consisting of three leading experts, including Prof. Yang Yang (University of California, Los Angeles), Prof. Yong Cao (South China University of Science and Technology) and Prof. Chihaya Adachi (Kyushu University) and the Executive Management Committee (EMC) consisting of all Co-PIs and had the day-to-day oversight of the project. With the assistance of the Technical Officer, the PC had executed the budget management as approved by EMC, closely monitored progress of projects, organized research symposia, workshops and EMC meetings, and the preparation of the Annual Progress Reports, Review Reports, and Completion Report for submission. The PC had also arranged five EMC meetings and formal scientific meetings annually.

7. Awards and Recognition

- 7.1 Have any research grants been awarded that are directly attributable to the results obtained from this project?

Team members have been successfully awarded major external grants arising from the results obtained from this TRS project, including Innovation and Technology Support Programme (ITSP) Tier 2 and Tier 3 projects, ITSP Guangdong-Hong Kong Technology Cooperation Funding Schemes, Guangdong Innovation Team Grant, Qatar National Research Fund, National Natural Science Foundation of China (NSFC) projects, RGC General Research Fund (GRF) and etc. Please refer to Appendix XIII for detailed research grants obtained.

- 7.2 Have any project team members participated as invited speakers in or organisers of international conferences as a result of this project?

Since the commencement of the TRS project, team members have delivered a total of **89** Invited Lectures related to this project in international/national conferences, among which **17** of these lectures were Plenary/Keynote/Award Lectures. Apart from the symposia and workshops organized, V.W.-W. Yam and A. Rogach were the organizers of the international workshop “Photonics of Functional Nanomaterials” that was held on 6–9 May 2013 at CityU. C.-S. Lee and C.-M. Che were also the Chair and Co-Chair of the MEMD2015 which was held on 5–8 January 2015 at CityU.

Furthermore, with the support from TRS, A. Rogach had organized a joint workshop on “CFP-NIM Workshop on Functional Photonics and Nanosystems” with the Nanosystems Initiative Munich on 27–29 May 2015 at CityU. Please refer to Section 6.8 for more details.

7.3 Have any project team members taken leadership positions in editorial boards, scientific and professional organisations?

V.W.-W Yam is the Associate Editor of *Inorg. Chem.*, Series Editor of the book series “*Energy & Environment Series*” published by RSC, member of the International Editorial Advisory Boards of *Angew. Chem.*, *Accounts Chem. Res.*, *J. Am. Chem. Soc.*, *ACS Nano*, *Chem*, *Chem. Sci.* and many others. C.-M. Che is member of International Editorial Advisory Board of *Coord. Chem. Rev.*, *Chem. Sci.*, *Chem. Commun.*, and many others. H.-S. Kwok is the Editor-in-Chief of *Chinese Journal of Liquid Crystals and Displays* and the Vice President of Society for Information Display. C.-S. Lee is member of RGC Physical Science Panel, the Editor-in-Chief of *Materials Today Energy*. K.-M. Ng is the Editor of *Comput. Chem. Eng.*, *Chem. Eng. Res. Des.* and *Curr. Opin. Chem. Eng.* C. Surya is the Chairman of IEEE Electron Devices Society Optoelectronics Technical Committee and member of the Editorial Board of *Nature: Sci. Rep.* F.-R. Zhu is Editor of *J. Mater. Sci. Mater. Electron.* and the Guest Editor of Special Issue on Organic Photovoltaics in *J. Mod. Optic.* W.-K. Chan is member of the Editorial Advisory Board of *ACS Appl. Mater. Inter.* G.-H. Chen is the Vice President of Hong Kong Institute of Science and the Associate Editor of *Eur. Phys. J. B.* A. Rogach is the Associate Editor of *ACS Nano*. B.-Z. Tang is the Editor in *Adv. Polym. Sci.*, the Associate Editor of *Polym. Chem.* and the Editor-in-Chief of *RSC Polym. Chem. Series* and many others. M. Wong is the Associate Editor of *J. Society of Information Display*. R.W.-Y. Wong is the Associate Editor of *J. Mater. Chem. C*, the Regional Editor of *J. Organomet. Chem.* Please refer to Appendix XIV for full list of leadership and invited editorships.



7.5 Other awards and recognitions as a result of this project (please specify):

We are delighted that our team members have received a number of prestigious awards and recognition as a result of this TRS project. V.W.-W. Yam has been named **Laureate of the 13th L’Oréal-UNESCO For Women in Science Awards** for her work on “Light-emitting materials and innovative ways of capturing solar energy”, and has been elected as **Foreign Member of Academia Europaea (The Academy of Europe)** in 2015 and as **Foreign Associate of the National Academy of Sciences, USA** in 2012. In addition, V.W.-W. Yam was awarded the **2012-13 Seaborg Memorial Lectureship** by UC Berkeley, **Doctorat Honoris Causa** by Université de Rennes 1, France, **Chinese Chemical Society (CCS)-Sinopec Chemistry Award** by CCS, **Julia S. and Edward C. Lee Memorial Lectureship (Lee Lectureship)** by University of Chicago, USA, **Royal Society of Chemistry (RSC) Ludwig Mond Award** by RSC, UK, **Japan Society of Coordination Chemistry International Award** from the Japan Society of Coordination Chemistry, the **Lavoisier Lectureship** from University Paris Diderot (Paris 7), France and the **APA Masuhara Lectureship Award** from Asian and Oceanian Photochemistry Association (APA). C.-M. Che has been elected as **Foreign Associate of the National Academy of Sciences, USA** in 2013 and was awarded the 2013 Centenary Prize by RSC, the **Ryoji Noyori Asian Chemical Editorial Society Award** in 2016, the **Lifetime Achievement Award in Organometallic Chemistry** by CCS in 2017 and **2016 HKU Faculty Knowledge Exchange Award**. B.-Z. Tang has been named **Laureate of the 27th Khwarizmi International Award** by the Iranian Organization for Science and Technology in 2014, and was awarded the Outstanding Contribution Award from Science China Chemistry in 2012, R.W.-Y. Wong was awarded the **2014 Lectureship Award for Asian and Oceanian Photochemist (Eikohsha Award)** by Japanese Photochemistry Association, the **Ho Leung Ho Lee Foundation Prize for Scientific and Technological Innovation in 2012**, and **Distinguished Lectureship Award** of the Chemical Society of Japan in 2012, the **Second**

Class State Natural Science Award from the State Council, P. R. China in 2013 and *Thomson Reuters Highly Cited Research Award* from 2014–16. One of the graduated student, Miss Haoning Tang, who was supervised by H.-S. Kwok, was awarded the *First Class Award in the Hong Kong University Students Technology Challenge Cup Competition 2015*, *Champion of HKUST ECE/EEIC Best Final Year Project Awards*, the *Winner of Varitronix 35th Anniversary Scholarship*, and *Silver Award of HKUST President's Cup Competition for Undergraduate Research and Innovation*.

8. Impacts

8.1 What are the current and expected impacts of the project on the long-term development of Hong Kong (social or economic development, e.g. patent, technology transfer, collaboration with external organisations, etc.)?

With the dedicated collaborative efforts by the team, the grand challenges on OLEDs and OPVs have been addressed and the deliverables were met with great success. Particularly, the team has disseminated new knowledge and technology in the form of **253** publications in high impact peer-reviewed journals (see Section 6.4) and exceptional performance on the development of PHOLED materials and new approaches for the realization of high-performance OPVs have been developed (see Section 6.1). In addition, a total of **26** patent applications have been filed, of which **14** patents have been successfully granted in US and national countries. Training of **~60** RPg students and young researchers has also been provided through the participation in the TRS project (see Section 6.6), helping nurture young scientists for Hong Kong and the region. Notably, strong collaborative links with top scientists and industrial partners have been established (see Section 9.3). While almost all commercially available OLED materials are iridium(III) phosphors where foreign companies, including Universal Display Corporation and Merck, holds most of the key patents and currently takes up more than 90 % of the OLED market, these Hong Kong-owned IP rights, patents and technological know-how can definitely overturn the business monopolies, as well as to motivate midstream and downstream R&D and commercialization activities in the Mainland, especially the Guangdong Province region. Noteworthy, *China's 13th Five-Year Plan has further emphasized six areas of emerging industries of strategic importance*, where it targets to make the added value of these strategic emerging industries accounting for 15 % of gross domestic product in China; *of which new display technology is in one of these six areas of emerging industries*.

The licensing is anticipated to realize next-generation OLED technologies with superior performance based on our proprietary platinum complexes. The establishment of "HKU-TCL Joint Laboratory for New Printable OLED Materials and Technology" will also enable the development of gold complexes to be further enhanced and expanded into newer dimensions, attaining greater heights and achievements in the near future.

8.2 Others (please specify): N/A

9. Sustainability of the Project

9.1 Whether there are new ideas evolved directly from this project?

The exceptional performance on the development of PHOLED materials and new approaches for the realization of high-performance OPVs obtained from this TRS project have evolved lots of new ideas for further development. Especially, new molecular design and strategies for the development of high-performance solution-processable OLED materials have been generated. New design strategies for reducing sublimation temperature and increasing radiative decay rate of phosphorescent metal complexes have also been established. Through efficient energy down-conversion, long-lived phosphorescent metal complexes can be used as sensitizer for enhancing the OLED performance. Rational design for air-stable and high-performance OLED materials based on inexpensive, earth-abundant metals have been generated. New molecular design approaches for utilizing exciplex emission and TADF properties have also been developed, overcoming the EQE limitation due to exciton spin statistics. Meanwhile, new ideas for the realization of high-performance OPVs and perovskite solar cells have been developed. These novel technologies can definitely provide a reliable and scalable method, paving the way for the

commercialization of OPVs and perovskite solar cells on large-area substrates. New idea on applying NIR-absorbing conjugated polymers for application in NIR phototransistors has been evolved. By using matrix-mediated nanowire networks of NIR-absorbing conjugate polymers, efficient NIR phototransistors with high charge transport mobility can be achieved. Industrial-competitive technology based on our pioneering BG structure that can greatly improve the performance of TFTs for AMOLEDs has been established.

9.2 Whether there are new projects evolved directly from this project?

As mentioned in Section 7.1, team members have been successfully awarded major external grants arising from the results obtained from this TRS project, including ITSP Tier 2 and Tier 3 projects, ITSP Guangdong-Hong Kong Technology Cooperation Funding Schemes, Guangdong Innovation Team Grant, Qatar National Research Fund, NSFC projects, RGC GRF and etc. In addition, the establishment of the "HKU-TCL Joint Laboratory for New Printable OLED Materials and Technology" as well as the "State Key Laboratory on Advanced Displays and Optoelectronics Technologies" can further sustain and enhance our excellence through world-class research towards the demonstration of superior technologies.

9.3 Whether there are new collaborations developed directly from this project?

The superior performance of our developed materials and technologies has attracted a lots of interests from industry. Strong collaborative links with renowned industrial partners.

9.4 Please give details on how much money and from which sources has been obtained/requested for the specific purpose of continuing the work started under this project.

Apart from the research grants mentioned in Section 7.1 that have been obtained to continue the works started under this project, the team has made tremendous efforts to sustain its activities. V.W.-W. Yam has been in collaboration with TCL Corporation for the establishment of the "HKU-TCL Joint Laboratory for New Printable OLED Materials and Technology".

Meanwhile, H.-S. Kwok has been successfully granted the establishment of the "State Key Laboratory on Advanced Displays and Optoelectronics Technologies", in collaboration with Sun Yat-Sen University. A full TRS proposal for renewal was submitted to the RGC in 2017 but was not funded despite excellent progress and favorable comments from the reviewers. Without further injection of funding support, the global competitiveness of the team will be greatly affected.