TRIS (8-HYDROXYQUINOLINATE) METALS FOR SOLUTION-PROCESSED ORGANIC SOLAR CELLS

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ABSTRACT

The simple fabrication process involving minimal material usage makes solution-processed organic solar cell (Courses) devices very attractive for harvesting solar energy. However, production of these devices on a commercial scale has been slow due to their relatively low power conversion efficiency and stability problems. It is expected that these obstacles will be surmounted in the future with rigorous studies actively being done in this field of research. Besides, a complete understanding of some basic electrical responses of these OSC devices has not been achieved yet. Consequently, seeking for interesting materials suitable for OSCs application and understanding the materials contribution are of great importance especially when strategies are targeted for the enhancement of OSCs. Tris (8-hydroxyquinolinate) metals (Mq3) are well known in the fabrication of stable organic light emitting diodes (OLEDs) and also for their unique optoelectronic properties. Very recently, tris (8-hydroxyquinolinate) aluminium (Alq3) prepared by thermal evaporation has been used as a buffer layer and dopant material to improve the performance of OSCs. However, its employment in solution-processed organic solar cells is still rare. Little attention has been paid on the behaviour of this material when applied in organic solar cells. Therefore, benefiting from the properties of Mq3 and easy fabrication process of solution-processed organic solar cell, the current thesis is focused on characterizing the OSCs related physical properties of tris (8-hydroxyquinolinate) gallium (Gaq3) and aluminium (Alq3) (as representatives of the Mq3 materials) and then applying them in solution-processed organic solar cells. The solution-processed OSC devices are based on ternary bulk heterojunction structure (three components blended all together) of dihexylisexithiophen/Mq3/methanofullerene (DH6T/Mq3/PCBM). The optoelectronics, spectroscopic, electrochemical, structural, morphological, and thermal properties of Mq3 materials are first investigated before

incorporating them into the photovoltaic active layers of the devices. From the analysis of physical properties of Mq3 materials as well as the assessment on the electrical characteristics of the devices, this work suggests that Mq3 can be a good candidate to be applied in solution-processed OSCs. The photovoltaic and electrical characteristics of the devices demonstrated that the photocurrent, open circuit voltage, and the performance of the devices have improved by approximately six times compared to the devices without Mq3 incorporation. The basic contribution of Mq3 materials for this improvement is believed to originate from the increase in the number of exciton generation and their dissociation into free charge carriers. This can be due to the enlarged area of the donor-acceptors boundaries between each of the DH6T/Mq3 and DH6T/PCBM moieties, thereby broadening the absorption of photons. Next, Mq3 incorporation can result in the stabilization of the mobility of the charge carriers within the DH6T donor and Mq3/PCBM acceptors producing a balanced transportation for the holes and electrons. The results indicated promising approaches for Mq3 materials to be applied in solution-processed OSCs as incorporation of Mq3 into the devices active layers considerably enhanced the overall performance and reproducibility of these devices.

ABSTRAK

Peranti sel suria organik (Courses) berasaskan larutan sangat menarik bagi menghasilkan tenaga suria kerana melibatkan penggunaan bahan yang minima melalui suatu proses pembuatan yang mudah. Walaubagaimanapun, penghasilan peranti ini pada skala komersial amat perlahan disebabkan kecekapan penukaran kuasa yang rendah secara relatifnya dan masalah kestabilan peranti. Tetapi, apabila kajian penyelidikan yang rapi dalam bidang ini digiatkan, dijangka halangan ini akan dapat diatasi pada masa depan. Pemahaman yang lengkap belum lagi dicapai bagi beberapa aspek asas elektrik dalam peranti OSC ini. Maka, pencarian bahan yang menarik dan sesuai bagi kegunaan OSC dan pemahaman terhadap peranan bahan, merupakan perkara penting terutamanya bagi mengatur strategi untuk meningkatkan prestasi peranti OSC. Logam tris (8-hydroxyquinolinate) (Mq3) dikenali ramai dalam penghasilan diod pemancar cahaya organik (OLED) dan juga sifat unik optoelektroniknya. Baru-baru ini, tris (8-hydroxyquinolinate) aluminium (Alq3) yang disediakan melalui kaedah pemendapan terma telah digunakan sebagai lapisan penampan dan bahan pendop bagi meningkatkan prestasi peranti OSC. Namun begitu, penggunaan bahan ini dalam sel suria organik berasaskan larutan masih jarang dijalankan. Hanya sedikit perhatian yang diberikan kepada sifat bahan ini apabila digunakan dalam sel suria organik. Oleh itu, berdasarkan kepada manfaat sifat bahan Mq3 dan proses pembuatan yang mudah untuk menghasilkan sel suria organik berasaskan larutan, tesis ini ditumpukan kepada mencirikan sifat-sifat fizikal berkaitan dengan OSC, yang menggunakan tris (8 hydroxyquinolinate) galium (Gaq3) dan Alq3, sebagai wakil daripada bahan Mq3, kemudian menggunakannya dalam pembuatan sel sel suria organik berasaskan larutan. Peranti dibuat berasaskan kepada simpang-hetero pukal ternari dihexylisexithiophen/Mq3/methanofullerene (DH6T/Mq3/PCBM). Sifat optoelektronik,

spektroskopi, elektro-kimia, struktur, morfologi, dan haba merupakan ciri awal yang dikaji sebelum bahan Mq3 digunakan sebagai lapisan aktif dalam peranti fotovoltaik. Hasil kajian ini mencadangkan bahawa Mq3 merupakan suatu bahan yang berpotensi untuk diaplikasikan dalam OSC berasaskan larutan, berdasar kepada analisa ciri fizikal bahan serta taksiran terhadap ciri elektrik peranti tersebut. Ciri fotovoltaik dan ciri elektrik peranti menunjukkan bahawa arus-foto, voltan litar-terbuka, dan prestasi keseluruhan peranti telah meningkat sebanyak kira-kira enam kali berbanding dengan peranti tanpa Mq3. Sumbangan asas bahan Mq3 kepada peningkatan ini, dipercayai berasal daripada peningkatan bilangan eksiton dan pemisahan eksiton menjadi pembawa cas bebas. Ini disebabkan kawasan sempadan penderma-penerima telah dibesarkan antara setiap komponen DH6T/Mq3 dan DH6T/PCBM, yang akhirnya menyebabkan pelebaran bagi serapan foton. Kemudian, penggunaan bahan Mq3 telah menyebabkan angkutan antara lohong dan elektron menjadi seimbang yang berpunca daripada kestabilan mobiliti pembawa cas di antara penderma DH6T dan penerima Mq3/PCBM. Keputusan kajian menunjukkan bahawa bahan Mq3 yang digunakan sebagai bahan lapisan aktif dalam peranti bagi pembuatan OSC berasaskan larutan, mampu memberi peningkatan bagi prestasi keseluruhan serta kebolehhasilan-semula peranti ini.

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RESEARCH PAPERS AND CONFERENCES

A. Papers Extracted from Thesis Contents

Muhammad, F. F., Abdul Hapip, A. I., & Sulaiman, K. (2010). Study of optoelectronic energy bands and molecular energy levels of tris (8-hydroxyquinolinate) gallium and aluminum organometallic materials from their spectroscopic and electrochemical analysis. *Journal of Organometallic Chemistry, 695*(23), 2526-2531.

Muhammad, F. F., & Sulaiman, K. (2011). Utilizing a simple and reliable method to investigate the optical functions of small molecular organic films - Alq3 and Gaq3 as examples. *Measurement, 44*(8), 1468-1474.

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Muhammad, F. F., & Sulaiman, K. (2011). Fabrication and characterization of solution-processed organic solar cells based on ternary bulk heterojunction of DH6T/Mq3/PCBM (M= Ga, Al). *Solar Energy Materials and Solar Cells,* (to be submitted).

B. Conferences Attended

National Physics Conference (*PERFIK2009*) - Malacca, Malaysia (Top 20 papers selection).

Fifth International Conference on Technological Advances of Thin Films and Surface Coatings (*ThinFilms2010*) - Harbin, China (Selection for ISI Journals).

Third International Conference on Functional Materials and Devices (*ICFMD2010*) - Terengganu, Malaysia (Gold Medal Achievement).

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I_{sc}	Short circuit current
η_{diff}	Exciton diffusion efficiency
η_{tc}	Hole-electron separation efficiency
η_{tr}	Carrier transport efficiency
η_{cc}	Charge collection efficiency
n_{∞}	Refractive indices at infinite wavelength
$\hat{n}$	Complex refractive index
$\hat{\varepsilon}$	Complex dielectric constant
ε_r	Real dielectric constant
ε_i	Imaginary dielectric constant
$\tan\delta$	Dissipation factor
α	Absorption coefficient
Φ_s	Volume fraction of solvent
T_{gs}	Glass transition temperature of solvent
T_{ms}	Melting temperature of solvent
λ	Wavelength
θ	Angle of diffraction
$\Delta\lambda_{1/2}$	Full width at half maximum (FWHM)
A	Absorbance
c	Velocity of light
d	Interplanner spacing
D-A	Donor-Acceptor
E	Photon energy
e	Electronic charge unit
E_{abs}	Absorption edge
E_g	Band gap energy
E_{red}	Reduction potential
FF	Fill factor
h	Planck's constant
h	Planck's constant

I_{max}	Current at maximum power
J	Current density
J_L	Photo-generated current density
J_o	Saturation current density
J_{sc}	Short circuit current density
k	Extinction coefficient
L	Mean free path of charge carriers
n	Refractive index
N_A	Acceptor density
N_C	Densities of states in the conduction band
N_V	Densities of states in the valence band
P_{in}	Input power, incident photon energy power
P_m, P_{max}	Maximum power
R_s	Series resistance
R_{sh}, R_p	Shunt resistance, parallel resistance (having same physical meaning)
S	Substrate refractive index
T	Transmittance
T_c	Crystalline temperature
T_g	Glass transition temperature
T_m	Melting temperature
T_{max}	Transmission maxima
T_{min}	Transmission minima
V_f	Diode potential barrier
V_{max}	Voltage at maximum power
V_{oc}	Open circuit voltage
η	Power conversion efficiency
μ	Charge carrier mobility
ν	Frequency

LIST OF ABBREVIATIONS

Al	Aluminum
Alq3	tris (8-hydroxyquinolinate) aluminum
AM	Air mass
Au	Gold
BCP	Bathocuproine
BHJ	Bulk heterojunction
BL	Bilayer
C_{60}	Buckminsterfullerene
Ca	Calcium
CB	Conduction band
CLB	Chlorobenzene
CNTs	carbon nanotubes
CS_2	Carbon disulphide
c-Si	crystalline silicon
CuPc	Copper phthalocyanine
CV	Cyclic voltammetry
DH6T	dihexyl-sexithiophene
DIW	distilled water
DMSO	Dimethyl sulphoxide
DSC	Differential scanning calorimetry
EA	Electron affinity
EBL	Exciton blocking layer
EM	Electron microscopy
EQE	External quantum efficiency
ETL	Electron transport layer
FESEM	Field emission scanning electron microscopy
FTIR	Fourier transform infrared
FWHM	Full width at half maximum
GaAs	gallium arsenide
Gaq3	tris (8-hydroxyquinolinate) gallium
GNDU	Ground unit
HCl	hydrochloric acid
HDI	Human Development Index

HOMO	higher occupied molecular orbital
IP	Ionization potential
IQE	Internal quantum efficiency
ITO	Indium-tin-oxide
kWh	kilowatt-hours
LiF	Lithium fluoride
LM	Light microscopy
LUMO	lower unoccupied molecular orbital
MEH-PPV	poly(2-methoxy-5(2'-ethyl) hexoxy-phenylenevinylene
Mg	Magnesium
MIM	metal-insulator-metal
MPP	maximum power point
Mq3	tris (8-hydroxyquinolinate) metals
OFET	Organic field effect transistor
OLED	organic light emitting diode
OLEDs	Organic light emitting diodes
P3HT	poly-3-hexylthiophene
PCBM	6,6-phenyl C_{61}-butyric acid methyl ester
PCPDTBT	poly[2,6-(4,4-bis-(2-ethylhexyl)-4H-cyclopenta[2,1-b;3,4-b']-dithiophen)-alt-4,7-(2,1,3-benzothiadiazole)]
PDTSTPD	thieno[3,4-c]pyrrole-4,6-dione and Dithieno[3,2-*b*:20,30-*d*]silole
PEDOT-PSS	poly(3,4-ethylenedioxythiophene):poly(4-styrenesulfonic) acid
PL	Photoluminescence
PPV	Poly(*p*-phenylenevinylene)
PTB4	fluorinated thieno[3,4-*b*] thiophene and benzodithiophene units
PTCBI	3,4,9,10-perylene tetracarboxylic-bisbenzimidazole
PV	photovoltaic
RES	Renewable Energy Sources
RS	Rayleigh scattering
SCE	Saturated calomel electrode
SCLC	Space charge limited current
SEM	Scanning electron microscopy
SHE	Standard hydrogen electrode (platinum)
SMU	Source measure unit
STC	Standard test condition

TBHJ	Ternary bulk heterojunction
TCAQ	tetracyanoanthraquino-dimethane
TFSCLC	Trap-filling space charge limited current
UV-Vis-NIR	Ultraviolet- Visible- Near Infrared
VB	Valence band
XRD	X-ray diffraction
ZnPc	Zincphthlocyanine
3D	Three dimensions
4T	Quarter-thiophene oligomer
6T	Sexi-thiophene oligomer