Optical Property Optimisation of Lead Sulphide Quantum Dots

A THESIS SUBMITTED BY

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DECLARATION

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The thesis was written in memorise of my grandfather. May he have peace in the other world.

ABSTRACT

To improve photovoltaic performance, lead sulphide quantum dots (PbS QDs) have been introduced in the device structure. To achieve enhanced performance by using PbS QDs, it is essential to gain better understanding on the particle size control and surface modification. The aim of this thesis is to gain better understanding of the particle size control, growth kinetics and surface modification of PbS particles to reach optimised properties for possible future applications.

The particle size modification and growth kinetics were studied by systematically varying reaction parameters of reaction time, temperature and reactant feed to synthesize a series of PbS QDs with different first exciton peaks. Studying the absorption wavelength and the full-width-at-half-maximum of photoluminescence spectra from different reaction times shows the three-stage growth process from nucleation through particle growth to final size saturation of the QDs. For each of these stages, the particle size and growth rate were found to be determined by reaction temperature and the stabilizer oleic acid concentrations. By analysing the change of absorption peaks with these parameters, the activation energy of the particle growth stage was calculated.

A novel surface ligand exchange approach was explored. By attaching the Pb onto the desired ligand functional groups, the formed 'atomic-ligand' could be readily used for solution phase ligand exchange. Tridentate poly(ethylene glycol) methyl ether (mPEG)

was used as a model hydrophilic ligand for PbS exchange. The modification of mPEG was confirmed, and the atomic ligand exchange was compared with the conventional ligand exchange by TEM imaging and quantum yield measurement. The results showed quantum yield enhancement of 715% from 2.7% to 22% and particle dispersity in polymer via atomic-ligand exchange compared with traditional method.

In conclusion, the thesis demonstrated detailed kinetics study of PbS quantum dots during different particle growth stages. For the first time, activation energy for PbS quantum dots particle growth was reported to be 28.8kJ/mol. A novel ligand exchange approached was first proposed and the quantum yield was observed to enhance by 715% comparing to conventional exchange method. The results from this study provided important results on PbS QDs synthesis and optimization, which would largely facilitate further studies on the understanding of principle PbS QDs growth dynamics and surface modification using various functional groups.

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LIST OF ABBREVIATIONS

AM 1.5	Air mass 1.5 distribution
BHJ	Bulk heterojunction
DMF	Dimethylformamide
DMSO	Dimethyl sulfoxide
DSSC	Dye-sensitized solar cell
DTPA	Diethylenetriaminepentaacetic acid
FF	Fill factor
FTO	Fluorine-doped tin oxide
FWHM	Full-width-at-half-maximum
HDA	Hexadecylamine
НОМО	Highest occupied molecular orbital
HSC	Heterojunction solar cell
ITO	Indium tin oxide
LUMO	Lowest unoccupied molecular orbital
MEH-PPV	Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene]
mPEG	Polyethylene glycol monomethyl ether
OA	Oleic acid
ODE	1-Octadecene
OSC	Organic solar cell
PC ₆₁ BM	[6,6]-phenyl-C61-butyric acid methyl ester
PCE	Power conversion efficiency
PEDOT:PSS	Poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate)
PV	Photovoltaic
QD	QD

- QE Quantum efficiency
- QY Quantum yield
- TEM Transmission electron microscope
- THF Tetrahydrofuran
- (TMS)₂S Hexamethyldisilathiane
- TOP Trioctylphospine
- TOPO Trioctylphosphine oxide