



**CHARACTERIZATION OF NICKEL
OXIDE QUANTUM DOTS AS HOLE
TRANSPORT MATERIAL IN
PEROVSKITE SOLAR CELLS**

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**MASTER OF SCIENCE
IN ELECTRONIC ENGINEERING**

2024



**Faculty of Electronics and Computer Technology
and Engineering**

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SUBATHRA A/P MUNIANDY

**A thesis submitted
in fulfillment of the requirements for the degree of Master of Science
in Electronic Engineering**

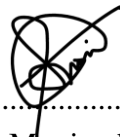
Faculty of Electronics and Computer Technology and Engineering

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

2024

DECLARATION

I declare that this thesis entitled “Characterization of Nickel Oxide Quantum Dots as Hole Transport Material in Perovskite Solar Cells” is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

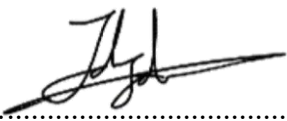
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APPROVAL

I hereby declare that I have read this thesis and in my opinion this thesis is sufficient in terms of scope and quality for the award of Master of Science in Electronic Engineering.

Signature	:..... 
Supervisor Name	:..... Dr. Muhammad Idzdihar Bin Idris
Date	:..... 19/01/2024

DEDICATION

This research paper is sincerely dedicated to my beloved mother and father who encouraged and inspired me in conducting this study. They have never left my side and gave me strength and hope throughout my study. They provide me with a great sense of enthusiasm and perseverance in continuing this research work. Without their love and assistance, this research would not have been made possible. Moreover, I dedicate this research paper to my research supervisor, Dr. Muhammad Idzdihar Bin Idris, and co-supervisor, Dr. Zul Atfyi Fauzan Bin Mohammed Napiah, who constantly provides guidance and support. I could not have undertaken this journey without them, who generously provided knowledge and expertise. Additionally, this research work endeavor would not have been possible without the generous support from Universiti Teknikal Malaysia Melaka (UTeM) for grant funding for the financial support throughout this project. I am also grateful to my friends, for their editing help, feedback sessions, and moral support. Lastly, I thanked Almighty God who gave me strength, wisdom, guidance, power of thinking, security, and competence and for giving me good health while doing this research work.

ABSTRACT

Perovskite solar cells (PSCs) have attracted the interest of researchers owing to their excellent light absorption, improved carrier mobility, high-power conversion efficiency and sustainable photovoltaic cell. PSCs have charge transport materials known as electron transporting layers (ETL) and hole transporting layers (HTL) that interact with the perovskite layer. The HTL is a fundamental component in PSC structures which are generally composed of a Spiro-OMeTAD and PEDOT:PSS material that has a drawback of a high cost, lengthy synthesis process, and insufficient long-term stability. Nickel oxide (NiO) as an inorganic HTL has been emphasized in PSCs owing to low cost, solution-based processing and the good band alignment as HTL in PSCs. Despite the extensive investigation of NiO, the importance of altering the pH of the precursor solution to improve the performance of HTL remains significant which have impact on the morphologies, crystal sizes, and textural qualities of the material. This project proposes a novel idea for introducing NiO quantum dots (QDs) in the HTL to achieve high efficiency and flexibility. The experimental study present the effect of the bottom and top layer of synthesized NiO at different pH values (9-12) using a spin-coating method annealed at 500 °C, 600 °C, and 700 °C. The in-depth characterization of the synthesized NiO was executed by XRD, UV-Vis spectroscopy, FTIR, AFM, SEM, TEM, PL, RF Impedance Analyzer, and four point-probe to investigate their structural, optical, element composition, surface roughness, morphological, quantum size, optical emission, dielectric constant, and resistivity properties. The performance of full fabricated PSC based on NiO or PEDOT:PSS/MAPbI₃/Graphene/ZnO/Ag were evaluated by current-voltage (I-V) curve. According to XRD findings, all the prominent diffraction peaks of NiO at 37.2°, 43.36°, 63.04°, and 75.51° were observed only in pH 11 at both layers. The SEM revealed the surface morphology of pH 11 have good coverage with less agglomeration of particles as compared to other pH values. The absorption spectrum of pH 11 was noticed in the UV region with band gap increasing from 3.45 to 3.64 for bottom layer and 3.42 to 3.47 for top layer. This research confirmed the quantum confinement effect obtained at pH 11 NiO through the reduction of crystallite size with higher bandgap energy. The impedance analyzer implies the layers of pH 11 NiO (700 °C) have higher dielectric constant (7.09 for bottom layer and 7.20 for top layer) with minimal polarization effect. A strong absorption peak were observed in FTIR analysis (400cm⁻¹ to 600cm⁻¹), ascribed to the presence of NiO vibration. TEM analysis revealed an average particle size of about 9.82 nm and 10.6 nm for the bottom and top layer of pH 11 NiO respectively. The observation of blue shifted PL emission bands that extended from 532 nm to 550 nm into the visible area proved that pH 11 NiO has high charge transfer resistance. The inverted PSC fabricated with bottom and top layer pH 11 NiO (700 °C) showed better performance than for the inverted PSC based on PEDOT:PSS. Based on these findings, NiO produced at pH 11 annealed at 700 °C exhibited promising characteristics, suitable for HTL in PSCs.

PENCIRIAN BINTIK KUANTUM NIKEL OKSIDA SEBAGAI BAHAN PENGANGKUTAN LUBANG DALAM SEL SURIA PEROVSKIT

ABSTRAK

Sel suria perovskit telah menarik minat penyelidik kerana penyerapan cahaya yang sangat baik, mobiliti pembawa yang lebih baik, kecekapan penukaran kuasa tinggi dan sel fotovoltai yang mampan. PSC mempunyai bahan pengangkut cas yang dikenali sebagai lapisan pengangkut elektron dan lapisan pengangkut lubang yang berinteraksi dengan lapisan perovskit. HTL ialah komponen asas dalam struktur PSC yang umumnya terdiri daripada bahan Spiro-OMeTAD dan PEDOT:PSS yang mempunyai kelemahan kos yang tinggi, proses sintesis yang panjang dan kestabilan jangka panjang yang tidak mencukupi. Nikel oksida (NiO) sebagai HTL tak organik telah ditekankan dalam PSC kerana kos rendah, pemprosesan berasaskan penyelesaian dan penjajaran jalur yang baik sebagai HTL dalam PSC. Walaupun penyiasatan meluas NiO, kepentingan mengubah pH larutan prekursor untuk meningkatkan prestasi HTL kekal penting yang mempunyai kesan ke atas morfologi, saiz kristal dan kualiti tekstur bahan. Projek ini mencadangkan idea baru untuk memperkenalkan bintik kuantum NiO dalam HTL untuk mencapai kecekapan dan fleksibiliti yang tinggi. Kajian eksperimen membentangkan kesan lapisan bawah dan atas NiO tersintesis pada nilai pH yang berbeza (9-12) menggunakan kaedah salutan putaran disepuh pada 500 °C, 600 °C dan 700 °C. Pencirian mendalam NiO yang disintesis telah dilaksanakan oleh XRD, spektroskopi UV-Vis, FTIR, AFM, SEM, TEM, PL, Penganalisis Impedans RF, dan pengesan empat bintik untuk menyiasat struktur, optik, komposisi unsur, kekasaran permukaannya, morfologi, saiz kuantum, pelepasan optik, pemalar dielektrik, dan sifat kerintangan. Prestasi PSC fabrikasi penuh berdasarkan NiO atau PEDOT:PSS/MAPbI₃/Grapene/ZnO/Ag telah dinilai oleh lengkung voltan semasa (I-V). Menurut penemuan XRD, semua puncak pembelauan ketara NiO pada 37.2°, 43.36°, 63.04°, dan 75.51° hanya diperhatikan dalam pH 11 pada kedua-dua lapisan. SEM mendedahkan morfologi permukaan pH 11 mempunyai liputan yang baik dengan kurang aglomerasi zarah berbanding dengan nilai pH lain. Spektrum penyerapan pH 11 telah diperhatikan di kawasan UV dengan jurang jalur meningkat daripada 3.45 kepada 3.64 untuk lapisan bawah dan 3.42 hingga 3.47 untuk lapisan atas. Penyelidikan ini mengesahkan kesan kurungan kuantum yang diperoleh pada pH 11 NiO melalui pengurangan saiz kristal dengan tenaga celah jalur yang lebih tinggi. Penganalisis impedans membayangkan lapisan pH 11 NiO (700 °C) mempunyai pemalar dielektrik yang lebih tinggi (7.09 untuk lapisan bawah dan 7.20 untuk lapisan atas) dengan kesan polarisasi yang minimum. Puncak penyerapan yang kuat telah diperhatikan dalam analisis FTIR (400cm⁻¹ hingga 600cm⁻¹), dikaitkan dengan kehadiran getaran NiO. Analisis TEM mendedahkan saiz zarah purata kira-kira 9.82 nm dan 10.6 nm untuk lapisan bawah dan atas pH 11 NiO masing-masing. Pemerhatian jalur pelepasan PL anjakan biru yang memanjang dari 532 nm hingga 550 nm ke kawasan yang boleh dilihat membuktikan bahawa pH 11 NiO mempunyai rintangan pemindahan cas yang tinggi. PSC terbalik yang direka dengan lapisan bawah dan atas pH 11 NiO (700 °C) menunjukkan prestasi yang lebih baik daripada PSC terbalik berdasarkan PEDOT:PSS. Berdasarkan penemuan ini, NiO yang dihasilkan pada pH 11 anil pada 700 °C mempamerkan ciri-ciri yang menjanjikan, sesuai untuk HTL dalam PSC.

ACKNOWLEDGEMENTS

First and foremost, I would like to take this opportunity to express my sincere acknowledgment to my supervisor Dr. Muhammad Idzdihar Bin Idris from the Faculty of Electronics and Computer Technology and Engineering Universiti Teknikal Malaysia Melaka (UTeM) for his essential supervision, support, and encouragement towards the completion of this thesis.

I would also like to express my greatest gratitude to Dr. Zul Atfyi Fauzan Bin Mohammed Napiah from the Faculty of Electronics and Computer Technology and Engineering, co-supervisor of this project for his advice and suggestions. Special thanks to UTeM for short-term grant funding for the financial support throughout this project. Additionally, this endeavor would not have been possible without the generous support from the Nano-Optoelectronics Research & Technology Laboratory (NOR Lab), School of Physics, Universiti Sains Malaysia (USM) and Functional Nanotechnology Device Laboratory, Institute of Nanoscience and Nanotechnology (ION2), Universiti Putra Malaysia (UPM) for the technical equipment used in my research.

Particularly, I would also like to express my deepest gratitude to Mr. Wahyudi, the technician from the Faculty of Electrical Technology and Engineering, Mr. Hairul Hisham, the technician from the material laboratory Faculty of Industrial and Manufacturing Technology and Engineering, Mr. Mahader the technician from the Faculty of Mechanical Technology and Engineering, Mr. Azmi, and Mr. Sufian, technicians from MiNe and microwave laboratory Faculty of Electronics and Computer Technology and Engineering, for their assistance and efforts in all the lab and analysis works.

Special thanks to all my peers, my mother, my beloved father, and my siblings for their moral support in completing this master's study. Lastly, thank you to everyone who had been to the crucial parts of the realization of this project.

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LIST OF ABBREVIATIONS

PV	-	Photovoltaic
Si	-	Silicon
c-Si	-	Crystalline silicon
a-Si	-	Amorphous silicon
PSCs	-	Perovskite solar cells
DSSC	-	Dye-sensitized solar cells
HTM	-	Hole transmission material
HTL	-	Hole transport layer
ETL	-	Electron transporting layer
ETM	-	Electron transport material
QDs	-	Quantum dots
IPCE	-	Incident photon to current conversion efficiency
GPVDM	-	General-Purpose Photovoltaic Device Mode
PCE	-	Power conversion efficiency
FF	-	Fill factor
XRD	-	X-ray diffraction spectrometer
SEM	-	Scanning electron microscope
UV-Vis	-	UV-visible spectroscopy
FTIR	-	Fourier-transform infrared spectroscopy
AFM	-	Atomic force microscope
TEM	-	Transmission electron microscopy
IRENA	-	International Renewable Energy Agency
PL	-	Photoluminescence
DC	-	Direct current
RMS	-	Root-mean-square
TCO	-	Transparent conductive oxide
OPV	-	Organic photovoltaic
JCPDS	-	Joint committee on powder diffraction standard
XPS	-	X-ray photoelectron spectroscopy
VB	-	Valence band
CB	-	Conduction band
SRH	-	Shockley-Read-Hall

HIA	-	Hybrid interfacial architecture
HOMO	-	Highest occupied molecular orbital
W-H	-	Williamson– Hall
FWHM	-	Full width at half maximum
3D	-	Three-dimensional
EDX, EDS	-	Energy dispersive x-ray spectroscopy
TN	-	True negative
VIM	-	Variation of information metric
Spiro-OMeTAD	-	2,2',7,7'-Tetrakis[N,N-di(4-methoxyphenyl)amino]-9,9'-spirobifluorene
PEDOT:PSS	-	poly(3,4-ethylenedioxythiophene) polystyrene sulfonate
NiO	-	Nickel oxide
ZnO	-	Zinc oxide
ITO	-	Indium-doped tin oxide
CuSCN	-	Copper(I) thiocyanate
CuI	-	Copper(I) iodide
Cu ₂ O	-	Copper(I) oxide
CuO	-	Copper(II) oxide
CoO	-	Cobalt(II) oxide
MAPbI ₃ , CH ₃ NH ₃ PbI ₃	-	Methylammonium lead iodide
Ag	-	Silver
SO ₂	-	Sulfur dioxide
NO _x	-	Nitrogen oxides
CdTe	-	Cadmium telluride
CIGS	-	Copper indium gallium diselenide
GaAs	-	Gallium arsenide
CaTiO ₃	-	Calcium titanate
Pb(II)	-	Lead(II)
Sn(II)	-	Tin(II)
I ⁻	-	Iodide
Br ⁻	-	Bromine
Cl ⁻	-	Chloride
Al	-	Aluminum
PbI ₂	-	Lead(II) iodide
MAI	-	Methylammonium iodide

TiO ₂	-	Titanium dioxide
In ₂ O ₃	-	Indium oxide
SnO ₂	-	Tin(IV) oxide
WO _x	-	Tungsten oxide
CNTs	-	Carbon nanotubes
MACl	-	Methylammonium chloride
AZO	-	Al into sol-gel ZnO
rGO	-	Reduced graphene oxide
FACs	-	Formamidinium mixed with cesium cations
MgO	-	Magnesium oxide
PCBM	-	Phenyl-C61-butyric acid methyl ester
GQDs	-	Graphene quantum dots
Li-TFSI	-	Lithium bis(trifluoromethane)sulfonimide
TBP	-	4-tert-butyl pyridine
P3HT	-	poly(3-hexylthiophene-2,5-diyl
PTAA	-	Poly(triarylamine)
LED	-	Light-emitting diodes
CuS	-	Copper sulfide
CuInS ₂	-	Copper indium sulfide
CuZnSnS ₄ , CZTS	-	Copper zinc tin sulfide
MoO _x	-	Molybdenum trioxide
VBM	-	Valence band maximum
MEA	-	Monoethanolamine
KOH	-	Potassium hydroxide
DEA	-	Diethanolamine
NaOH	-	Sodium hydroxide
PVA	-	Polyvinyl alcohol
H ₃ O ⁺	-	Hydronium ion
OH	-	Hydroxide
PEG	-	Polyethylene Glycol
PVP	-	Polyvinylpyrrolidone
NiCl ₂	-	Nickel chloride
FESEM	-	Field emission electron microscope
GO	-	Graphene oxide
FTO	-	Fluorine-doped tin oxide

Ni(OH) ₂	-	Nickel hydroxide
Cs	-	Cesium
BCP	-	Bathocuproine
C ₆₀	-	Buckminsterfullerene
ZrAcac	-	Zirconium acetylacetonate
DMF	-	N,N dimethylformamide
DMSO	-	Dimethyl sulfoxide
EC	-	Ethyl cellulose
C ₁₀ H ₁₈ O	-	α-terpineol
IPA, C ₃ H ₈ O	-	Isopropyl alcohol
C ₂ H ₅ OH	-	Ethanol
DI	-	Deionized water
CoO	-	Cobalt(II) oxide
Ni(CH ₃ COO) ₂ ·4(H ₂ O)	-	Nickel acetate tetrahydrate
SiC	-	Silicon carbide
GaN	-	Gallium nitride
MOSFET	-	Metal–oxide semiconductor field-effect transistor
Pt	-	Platinum
Ni	-	Nickel
C	-	Carbon
O	-	Oxygen
ACQ	-	Aggregation caused quenching
AIE	-	Aggregation-induced emission
DLE	-	Deep-level-emission
V _o	-	Oxygen vacancies
Sb-	-	Antimony
Bi-	-	Bismuth
Ge-	-	Germanium
Cu-	-	Copper
SILAR	-	Successive ionic layer adsorption and reaction
KBr	-	Potassium Bromide
CO ₂	-	Carbon dioxide
H ₂ O	-	Water