THEORETICAL AND EXPERIMENTAL INVESTIGATIONS OF ZINC OXIDE NANOSTRUCTURES AS PHOTOELECTRODE FOR SOLAR CELL APPLICATION

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I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang Al-Sultan Abdullah or any other institutions.

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ABSTRAK

Para saintis telah berminat dengan sel suria selama bertahun-tahun kerana keupayaannya menjana tenaga elektrik daripada cahaya matahari. Salah satu perkembangan terkini yang menjanjikan dalam bidang ini ialah sel suria titik kuantum (QDSC) yang berpotensi menggantikan sel suria terpeka pewarna (DSSC) kerana struktur dan persamaan perantinya yang agak ringkas. Dalam QDSC, kesan fotovoltaik (PV) berlaku pada antara muka antara elektrolit redoks dan titik kuantum (QD) terkonjugasi semikonduktor oksida logam (MOS) jurang jalur lebar, bukannya molekul pewarna. QD juga mempunyai beberapa kelebihan berbanding pewarna organik, termasuk sifat elektrik dan optik yang lebih baik kerana beberapa sebab, iaitu: (i) jurang jalur boleh tala yang bergantung kepada saiz QD, (ii) keratan rentas penyerapan yang lebih besar, dan (iii) keupayaan untuk menghasilkan penyerapan foton tunggal (MEG) daripada penyerapan satu-foton. Sejak kebelakangan ini, penyelidik hanya tertumpu pada komponen fluorofor dan elektrolit dalam sel suria berbanding fotoelektrod. Penyelidikan ini bertujuan untuk (i) mefabrikasi nanozarah (NP) ZnO menggunakan kaedah penyejatan haba tervakum (TE) dan mengkaji sifat opto-elektroniknya, (ii) mengkaji kesan kitaran penyejatan terhadap sifat NP ZnO yang difabrikasikan, (iii) membina dan mengesahkan model kluster realistik ZnO NPs menggunakan teori fungsian kepadatan (DFT) *ab-initio*. Tekanan TE 5×10^{-4} Torr dan 5×10^{-5} Torr boleh menunjukkan beberapa kenaikan iaitu (i) jurang jalur 2.58 eV dan 2.80 eV, dan (ii) kecekapan PV peranti ZnO/PbS/ polimer karbonatoksiletil selulosa dan polyvinyl alkohol (CMC-PVA) 0.00181% dan 0.99%. Kitaran penyejatan yang berbeza digunakan iaitu, kitaran satu, dua, dan tiga penyejatan akan mengubah julat saiz taburan nanosfera ZnO (berkurangan apabila bertambahnya kitaran penyejatan) dan jurang jalur nanosfera ZnO (meningkat apabila bertambahnya kitaran penyejatan). Geometri struktur ZnO telah dikenalpasti iaitu, $(ZnO)_3$, $(ZnO)_5$, $(ZnO)_6$, $(ZnO)_{16}$, $(ZnO)_{24}$, dan $(ZnO)_{30}$ dengan saiz 1.788 nm, 2.459 nm, 2.651 nm, 4.997 nm, 6.543 nm, dan 7.529 nm. Penjajaran aras tenaga yang melibatkan orbit molekul terendah yang diduduki oleh elektron–elektron di dalam CdSe ($LUMO_{CdSe}$) dan $(ZnO)_{30}$ dihipotesiskan akan memenuhi syarat iaitu, (i) suntikan elektron daripada CdSe ke ZnO ($LUMO_{CdSe} > LUMO_{ZnO}$), dan (iii) pengangkutan elektron yang cekap ($LUMO_{ZnO} < LUMO_{CdSe}$). Puncak eksitonik pertama $(ZnO)_{30}$ adalah selari dengan baik dengan filem nipis ZnO, oleh itu dihipotesiskan bahawa kelompok realistik $(ZnO)_{30}$ boleh difabrikasikan. Kesimpulannya, $(ZnO)_{30}$ nano-sfera boleh difabrikasikan menggunakan TE dengan tekanan fabrikasi 5×10^{-5} Torr dan tiga kitaran penyejatan untuk penyerapan filem nipis yang lebih baik.

ABSTRACT

Scientists have been interested in solar cells for many years due to its ability to generate electrical energy from sunlight. One of the promising recent developments in this field is the quantum dots solar cell (QDSC) that can potentially replace dye sensitized solar cell (DSSC) due to their relatively simpler device structure and similarity. In QDSCs, the photovoltaic (PV) effect occurs at the interface between a redox electrolyte and the quantum dot (QD) conjugated wide bandgap metal oxide semiconductor (MOS), instead of dye molecules. QDs also have several advantages over organic dyes, including better electrical and optical properties due to several reasons, which are: (i) QD size-dependent tuneable bandgap, (ii) larger absorption cross-section, and (iii) ability to produce multiple excitons generation (MEG) from the absorption of a single-photon. In recent years, researchers have been focusing only on fluorophores and electrolytes in solar cells rather than on the photoelectrode. This research aims to (i) fabricate ZnO nanoparticles (NPs) using the thermal evaporation (TE) method and study their opto-electronic properties, (ii) study the effect of evaporation cycles to the properties of fabricated ZnO NPs, and (iii) build and validate realistic cluster models of ZnO NPs using ab-initio density functional theory (DFT) calculations. The TE pressure of 5×10^{-4} Torr and 5×10^{-5} Torr exhibited increments in: (i) bandgap of 2.58 eV and 2.80 eV, and (ii) PV efficiency of device ZnO/PbS/Carboxymethyl cellulose and polyvinyl alcohol (CMC-PVA) of 0.00181% and 0.99%, respectively. The different evaporation cycles i.e., one-, two-, and three-evaporation cycles caused variations in the size range of distribution of ZnO nanosphere (decreasing upon increment of evaporation cycles), and bandgap of ZnO nanosphere (increasing upon increment of evaporation cycles). The structural geometries of ZnO were identified viz., $(ZnO)_3$, $(ZnO)_5$, $(ZnO)_6$, $(ZnO)_{16}$, $(ZnO)_{24}$, and $(ZnO)_{30}$ with the size of 1.788 nm, 2.459 nm, 2.651 nm, 4.997 nm, 6.543 nm, and 7.529 nm respectively. The energy level alignment which involves the lowest occupied molecular orbitals of CdSe ($LUMO_{CdSe}$) and $(ZnO)_{30}$ is hypothesized to be able to satisfy the demand of i.e., (i) electron injection from CdSe to the ZnO ($LUMO_{CdSe} > LUMO_{ZnO}$), and (iii) efficient electron transport ($LUMO_{ZnO} < LUMO_{CdSe}$). The first excitonic peak of $(ZnO)_{30}$ is in a good agreement with that of the ZnO thin film, thus it is hypothesized that the $(ZnO)_{30}$ realistic cluster could be fabricated. In conclusion, the $(ZnO)_{30}$ nano-sphere could be fabricated using TE with the fabrication pressure of 5×10^{-5} Torr and three-evaporation cycle for a better adsorption of thin film.

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