

**FUNDAMENTAL STUDY OF DC AND AC  
ELECTRICAL RESPONSE ON A  
HETEROJUNCTION DEVICE (AL/ZNO-  
PTAA/METAL) FOR OPTOELECTRONIC  
APPLICATION**

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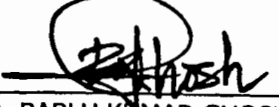
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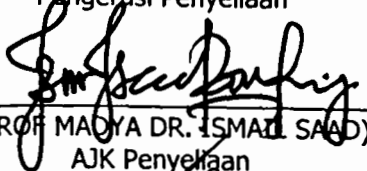
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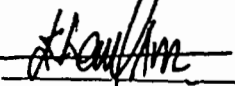
  
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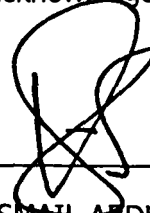
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24<sup>th</sup> July 2019



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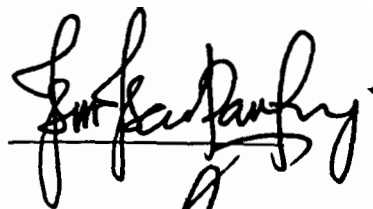
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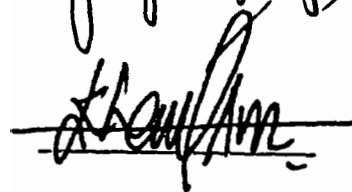


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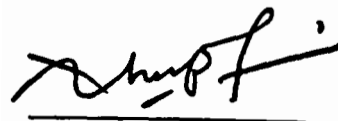
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## ABSTRACT

The combination of organic and inorganic semiconductor materials has unlocked new achievements in the performance of semiconductor devices. In the recent years, technology of hybrid material system consisting of inorganic-organic semiconductors has sufficiently evolved and is becoming popular among researchers. The inorganic semiconductors form huge crystalline networks with strong covalent or ionic bond when deposited onto a substrate layer. The organic semiconductors with smaller molecules, layers itself onto the inorganic film produces a strong grip at the interface. This property leads to a higher electrons and holes carrier mobility in the device. However, all organic based device has been limited by low speed of constituent organic devices and inorganic devices has several disadvantages for high frequency rectification due to their poor current density and low rectification ratio. Therefore, this research focuses on a hybrid inorganic-organic diode composed of two materials with different structural and optical properties. The diode is combination of Zinc Oxide (ZnO) and Polytriarylamine (PTAA) conjugated polymer acting as n-type and p-type semiconductor materials, respectively. Morphology ZnO and PTAA thin films were fabricated by varying the spin speed and temperature using combination of RF sputtering and spin coating techniques repeatedly. The hybrid diode was design by forming a PTAA-ZnO layer in between indium tin oxide (ITO) and aluminium electrode (Al) based on spin speed of 1000 and 2000 rpm with annealing temperature of 100 to 150°C. The X-ray diffraction pattern for ZnO clearly showed a broad diffraction peak at 34.2°. Meanwhile, the PTAA film depicted a broad peak between the angles of 23° to 25°. Besides that, both materials presented a transmittance about 80% and above in the wavelength range of 370 to 850 nm using UV-vis spectrometer. The surface morphological of both thin films was observed using an advanced material microscope (HIROX). The images of ZnO-PTAA thin films indicated the layer of PTAA change colour from light blue to yellowish as the annealing temperature increases, which shows a degradation over high temperature. Furthermore, as the spin speed increases, the distribution is homogenously distributed on the surface of ZnO. The current-voltage (I-V) characteristics were measured using Keithley 2400 source meter from -4.0 to 4.0 V. The turn-on voltages are varying from 1.4 V to 2.5 V depending on crystallinity and homogenous distribution of thin films. Lastly, the frequency dependent electrical response was measured using precision LCR meter in the frequency range up to 100 KHz with various bias voltages of 1.0 to 5.0 V. The results revealed that capacitance was independent towards the high frequency level of 10 Hz to 100 KHz but conductance and series resistance are strongly dependent on the frequency and bias voltage. In conclusion, combination of ZnO and PTAA semiconductors for a heterojunction device showed unique structural and optical properties and significantly improve the electrical properties due to the extended framework of the PTAA with fusion of high crystallinity structure of ZnO on thin film uniformity and performance stability.



## ABSTRAK

### **KAJIAN ASAS TINDAK BALAS ELEKTRIK DC DAN AC PADA PERANTI HETERO-SIMPANG (AL/ZNO-PTAA/ LOGAM) UNTUK APLIKASI OPTOELEKTRONIK**

Gabungan bahan semikonduktor organik dan bukan organik telah memberi ruang untuk pencapaian baru dalam prestasi peranti semikonduktor. Dalam tahun-tahun kebelakangan ini, teknologi sistem bahan hibrid yang terdiri daripada semikonduktor organik dan bukan organik telah cukup berkembang dan menjadi popular dalam kalangan penyelidik. Semikonduktor bukan organik membentuk rangkaian kristal yang besar dengan ikatan kovalen atau ionik yang kuat apabila dimendapkan ke lapisan substrat. Semikonduktor organik dengan molekul yang lebih kecil, membentuk lapisan pada filem bukan organik dan menghasilkan cengkaman yang kukuh di antara muka lapisan tersebut. Sifat ini membawa kepada mobiliti pengangkut elektron dan lubang yang lebih tinggi dalam peranti. Walau bagaimanapun, semua peranti berasaskan bahan organik adalah terbatas dengan kadar penghasilan yang mengambil masa dan peranti bukan organik mempunyai beberapa kelemahan apabila berada dalam keadaan berfrekuensi tinggi disebabkan ketumpatan arus yang lemah dan rendah. Oleh itu, kajian ini memfokuskan pada diod hibrid organik dan bukan organik yang terdiri daripada dua bahan yang mempunyai sifat struktur dan optik yang berbeza. Diod ini ialah gabungan Zink Oksida (ZnO) dan polimer berkonjugat Politriarilamin (PTAA) yang masing-masing bertindak sebagai bahan jenis-n dan bahan semikonduktor jenis-p. Filem-filem nipis morfologi ZnO dan PTAA dibuat dengan mengubah kelajuan putaran dan suhu dengan menggunakan gabungan teknik percikan berfrekuensi radio dan teknik salutan berputar. Diod hibrid ini direkabentuk dengan membentuk lapisan PTAA-ZnO di antara indium tin oksida (ITO) dan elektrod aluminium (Al) pada kelajuan putaran 1000 dan 2000 rpm dengan suhu penyepuhlingapan 100° hingga 150° C. Corak difraksi sinar-X untuk ZnO jelas menunjukkan puncak difraksi yang luas pada 34.2°. Sementara itu, filem PTAA menggambarkan puncak luas antara sudut 23° hingga 25°. Selain itu, kedua-dua bahan membentangkan penembusan kira-kira 80% dan keatas dalam julat panjang gelombang 370 hingga 850 nm menggunakan spektrometer UV-vis. Morfologi permukaan kedua-dua filem nipis diperhatikan menggunakan mikroskop bahan canggih (HIROX). Gambar-gambar filem nipis ZnO-PTAA menunjukkan lapisan PTAA bertukar menjadi kekuningan apabila suhu penyepuhlingapan bertambah. Hal ini menunjukkan kemerosotan pada suhu tinggi. Selain itu, apabila kelajuan putaran meningkat, taburan diedarkan secara sekata pada permukaan ZnO. Ciri-ciri arus-voltan (I-V) diukur dengan menggunakan meter sumber Keithley 2400 dari -4.0 ke 4.0 V. Voltan putaran adalah berbeza daripada 1.4 hingga 2.5 V bergantung kepada pengikatan kristal dan pengedaran filem nipis. Akhir sekali, tindak balas elektrik yang bergantung kepada frekuensi diukur dengan menggunakan meter kejitaan LCR dalam frekuensi sehingga 100 kHz dengan pelbagai voltan bias 1.0 hingga 5.0 V. Hasilnya menunjukkan kapasitan dan rintangan siri tidak bergantung kepada frekuensi tinggi dari 10 hingga 100 kHz tetapi konduktansinya sangat bergantung kepada kekerapan dan voltan bias. Sebagai kesimpulan, hasil gabungan semikonduktor ZnO dan PTAA untuk peranti simpang hetero menunjukkan sifat struktur dan optik yang unik dan membuktikan bahawa sifat elektrik meningkat dengan ketara kerana struktur PTAA yang bersesuaian dengan gabungan struktur kristalografi ZnO yang tinggi pada keseragaman filem nipis dan kestabilan prestasi.

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## LIST OF ABBREVIATIONS

RFID	Radio Frequency Identification
TFTs	Thin Film Transistor
RF	Radio Frequency
IV	Current - voltage
ITO	Indium Tin Oxide
RPM	Revolution per minute
AC	Alternating Current
DC	Direct Current
EV	valence band
$E_c$	conduction band
$E_g$	Energy band gap
HOMO	Highest Occupied Molecular Orbital
LUMO	Lowest Un-occupied Molecular Orbital
FWHM	Full Width Half Maximum
sccm	Standard Cubic Centimetre Per Minute
eV	electron volt
°C	Celsius
K	Kelvin
cm	Centimetre
V	Volt
s	Second
$\Omega$	Ohm
Hz	Hertz



## LIST OF SYMBOLS

$\pi$	pi
$w$	spin speed
$\beta_0$	initial kinematic viscosity
$j$	current density
%	percentage
A	Ampere
W	Watt
$\lambda$	wavelength
$\theta$	angle
$d$	Spacing between plane
$D$	average grain size
$\beta$	Width of FWHM
$I_i$	prefactor current
$n$	ideality factor
$\phi_b$	barrier height
$E_g$	band gap energy
$X_s$	electron affinity of the semiconductor
$Y$	admittance
$Z$	impedance
$G$	conductance
$w$	angular frequency
$f$	frequency
$C$	capacitance
$R_s$	series resistance
°	degree
Å	Angstrom
$\sigma$	electrical conductivity

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# CHAPTER 1

## INTRODUCTION

### 1.1 BACKGROUND OF STUDY

Organic- inorganic heterojunctions have a promising future because of the variety of optical and electrical properties, which make them technologically useful for optoelectronics applications. The main idea in developing hybrid devices was to take advantages of the best properties of each component that forms a heterojunction, trying to decrease or eliminate their drawbacks getting in an ideal way of synergic effect that results in the development of the new systems with new properties. Inorganic materials offer a broader absorption of spectrum compared to the organic semiconductors. Its characteristic resemblance as irreplaceable electronic, optical and biocompatible properties which result in improvement of response and sensitivity of the TFTs (Mohamad *et al.*, 2015). Wide range resistance, high optical transparency in the visible light region and wide bandgap energy enhanced the application of oxide material for optoelectronics application (Afishah *et al.*, 2013).

Nowdays organics semiconductor are widely accepted as an active semiconductor materials based on their unique characteristic such as light weight, simple structure, more flexibility, and more versatile than inorganics device (Mohamad *et al.*, 2015). Organic semiconductors is considered as active materials due to van der waals intermolecular forces and offer wide range of optical



properties. Combining such organics semiconductors with inorganics semiconductors of wide optical bandgap is promising for optoelectronics applications such as radio frequency identification (RFID) tags (Cantatore *et al.*, 2006; Gowrisanker *et al.*, 2009). High frequency RFID tags require a diode to operate at 13.56 MHz frequency with maximum current density and a sufficiently large reverse breakdown voltage and low forward turn-on voltage (Finkenzeller, 2011; Pal *et al.*, 2008). The rectification ratio should be in the range 100 to 1000 (Finkenzeller, 2011). A rectifier consists of diode and capacitors. Diodes can be classified into p-n junction, transistors and Schottky diodes. A p-n junction diode integrates p-type and n-type semiconductors.

Organic thin films have been used as a p-type semiconductor since it is an effective hole transport materials with relatively high charge carrier mobility (Hunter *et al.*, 2014). Several publications are also reported that incorporating organic materials such as poly(trialamine) (PTAA) in hetero-structure with different inorganic semiconductors showed promising electrical characteristics (Heo *et al.*, 2013; Logan *et al.*, 2015). Recently, PTAA-based devices was tested to be air stable under different environment conditions which make it favor for hybrid radio frequency identification (RFID) operates under ambient conditions (Lau *et al.*, 2013). On the other hand, undoped zinc oxide (ZnO) thin films have an n-type conduction and a high resistivity due to low carrier concentration with a wide bandgap energy (3.3 eV), a large exciton binding energy (60 meV) and large saturation velocity (100 cm/s), which indicated that ZnO-based devices would be better for high frequency applications, thus making it ideal for optoelectronic applications. (Lee *et al.*, 2010; Prepelita *et al.*, 2010). Despite the extensive research work on the subject, stable p-type ZnO is difficult to manufacture, thus hindering the fabrication of ZnO p-n homojunctions (Fan *et al.*, 2013). Several hetero-junction organic-inorganic devices have been proposed and recently fabrication p-n pentacene/ZnO diode producing uniform layers under ambient condition without involving high energy processes with nonlinear characteristics similar to that of inorganic p-n junction diode was reported (Dzulfahmi *et al.*, 2015)

Most reported organic-inorganic p-n junction diodes have been extensively investigated for their carrier injection mechanism. The inorganic semiconductor forms an extended framework bound by strong covalent or ionic interactions to provide high carrier mobility. Meanwhile, the organic semiconductor facilitates the self-assembly of these materials, enabling heterojunctions to be deposited by the simple, low-temperature as the organic materials. Organic-inorganic diodes with high current density with low operating voltage require a substantial interfacial contact between thin films to operate at high frequency rectification under ambient environment conditions. For high speed and high current density diodes, the interface resistance should be very low at forward bias, low conductance in reverse bias and a high reverse breakdown voltage. A diode only requires electrons and holes flow on nanometer scale lengths from electrodes to the recombination region, current density and speed can be expected to be higher compared to homojunction devices when numerous grain boundaries and defect sites are minimized at the organic and inorganic interface during the fabrication process.

## 1.2 Problem Statement

Nowdays, the semiconductor technology is becoming one of the most important industrial revolution towards the world demand on clean and green technology that not only save for the environment but also less cost manufacturing. Organic and inorganic based electronics device had temporarily helped the industry to uphold the policy but however the progress in all-organic-based devices has been limited by the low speed of the constituent organic devices. Meanwhile, inorganic-based devices have several disadvantages for high frequency rectification due to their poor current density, very low rectification ratio and relatively lower reverse breakdown voltage. Thus combination organic-inorganic might overcome this drawback using the versatility of the chemical structure of organic with the established properties of inorganic. In spite of many advantages of this combination organic-inorganic, there are few drawbacks which have to be overcome in order to meet application requirement that comparable to the conventional silicon devices. The relative

instability and poor electrical performance are among the drawbacks with due to interface traps. As much works focusing on the design and configuration of organic-inorganic heterojunctions such as PTAA-ZnO combination to increase the energy efficiency, less attention on analysis of the dominating conduction mechanism, i.e. direct current (DC) and alternating current (AC) electrical responses, which is very important for the operation of high quality optoelectronic applications.

The main idea in developing organic-inorganic devices was to take advantages of the best properties of each material that forms a heterojunction and trying to decrease or eliminate their drawbacks getting in an ideal way a synergic effect that results in the new systems with new properties. However, little is known about the PTAA-ZnO for their intrinsic interaction at the interface as well as its applications. Charge transport characteristics are key attributes that have important consequence for the device electrical performance. Thus this project is focusing to elucidate the frequency dependent impedance such as frequency dependent conductance, capacitance and current-voltage characteristics using impedance or AC analysis.

### **1.3 Research Objective**

The main aim of this research is to fundamentally investigate the frequency dependent electrical responses on organic-inorganic Schottky diode focused on the optoelectronics application. This research focusses on the following objectives:

- To investigate the effect of annealing temperature on the electrical responses of a heterojunction PTAA-ZnO Schottky diode.
- To determine the dc electrical characteristics of a heterojunction PTAA-ZnO Schottky diode by current-voltage relationship
- To model equivalent circuit based on the current-voltage characteristics of a heterojunction PTAA-ZnO Schottky diode using frequency dependent electrical responses.

## 1.4 Research Scope

This research focus on design, fabrication and characterization, which conducted to determine the optimum deposition speed and annealing temperature of PTAA during deposition that will optimize the DC electrical characteristics and AC frequency dependent impedance characteristics of the hybrid Inorganic-organic diode. In the process of diode fabrication, radio frequency (RF) sputtering, spin coating technique and thermal evaporation was used as the inorganic-organic semiconductor material deposition technique to form thin film or layer. RF sputtering and spin coater enable the thin film to be fabricate uniformly. Thus, RF sputtering is for the inorganic material with the aid of spin coating technique for organic materials which easily to be dissolved in solvent. Besides that, all experiments to fabricate thin films and hybrid diode were conducted in a controlled environment by minimizing the exposure of the fabricated diode with ambient environment. The DC and AC measurement setup is required to conduct electrical and frequency-dependent impedance of hybrid diodes.

## 1.5 Thesis Outline

The thesis contains five chapters. Chapter 1 is an introduction that gives a brief explanation of the technology that involving inorganic, organic semiconductor, diode and what is the limitation in achieving high efficiency inorganic-organic diode. The research aim, objective scope of this thesis is presented in this chapter.

Chapter 2 gives a brief explanation about the basic theory behind inorganic-organic semiconductor. A brief review on previous research is written to show the various materials that are used for the development of inorganic-organic devices.

Experimental steps are explained in chapter 3. This chapter explains the details on pre- and post-processing procedures and steps in order to form a thin film or layer and fabricate the hybrid devices. A physical characterization method of determining thin film properties such as X-Ray diffraction and SEM is also briefly explained, while experimental setup for the DC and AC measurement using an LCR



meter and computer-controlled current-voltage measurement were also explained to determine the electrical properties of hybrid inorganic-organic devices.

Chapter 4 focusing on the results that obtains from the experiment. The characterization has been selected which is the frequency dependent electrical characteristics and the DC current-voltage (I-V) characteristics of the diode device.

Chapter 5 presenting the summary of the findings, conclusion and also recommendation. All the result of this work is summarized. By the thesis data, a future work regarding the thin film performance is described.

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