

**FORMATION AND CHARACTERIZATION OF THIN FILM FROM THE
EXTRACTION OF TOMATO FOR UV PHOTODETECTOR**

by

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LIST OF ABBREVIATIONS

(COOH) ₂ .2H ₂ O	Oxalic acid.
AFM	Atomic Force Microscopy.
BAIq	Tris-(8-hydroxyquinoline) aluminium.
BCP	Bathocuproine.
BHJ	Bulk Heterojunction.
CMOS	Complementary metal–oxide–semiconductor structure.
CuBB	Cu(1,2-bis(diphenylphosphino)benzene) (bathocuproine).
D*	Detectivity.
E-Waste	Electronic Waste.
FESEM	Field Emission Scanning Electron Microscope
FET	Field effect transistor.
FTIR	Fourier Transformed Infrared spectroscopy.
ITO	Indium–tin-oxide.
IR	Infrared-Red.
J	Current Density.
m-MTDATA	Methyl-pheny(phenyl)amino]triphenylamine.
MSM	Metal–semiconductor-metal structure.
MOS	Metal-oxide-semiconductor structure.
n	Refractive Index.
NDI	N,N'-Bis (phenylmethyl) naphthalene.
NTCDI	Naphthalene diimide.
NPB	N-bis(1-naphthyl)-(1,1(-benzidine)-4,4-diamine.

NPN	Double junction formed by sandwiching a thin layer of p-type materials between two layers of n-type material.
NSN	Bis (4-(4,6-diphenyl-1,3,5-triazine-2-yl) phenyl) diphenylsilane.
PBD	(2-(4-tertbutylphenyl)-5-(4-biphenyl)-1, 3, 4 oxadiazole).
PEDOT	Poly(3,4-ethylenedioxythiophene).
PFP	2,7-bis(30,50-diphenylphenyl)-9,9-diphenylfluorene.
PFH	Poly (9,9-dihexylfluorene-2,7-diyl).
PHJ	Planar Heterojunction.
P(i)N	P-type intrinsic N-type structure.
PNP	Double junction formed by sandwiching a thin layer of n-type materials between two layers of p-type material.
PSS	Poly (styrenesulfonate).
PVK	Poly(N-vinylcarbazole).
R	Responsivity.
THF	Tetrahydrofuran.
TGA	Thermogravimetric analyser.
TPBi	1,3,5-tris(Nphenylbenzimidazol-2-yl)-benzene.
UV	Ultraviolet.
VUV	Vacuum Ultraviolet.
WBG	Wide-Band Gap.

LIST OF SYMBOLS

%	Percentage
<	Less than
>	More than
D*	Detectivity
°C	Degree Celsius
°C/min	Degree Celsius per minute
A/W	Responsivity
A/cm ²	Current density
cm ⁻¹	Wavenumber
η	Quantum efficiency
h	Hour
L	Litre
m	Meter
min	Minute
τ	Times rate
mm	Millimetre
rmp	Voltage rate per second
wt %	Weight percent
λ	Wave length
V	Voltage
nm	Nano meter
g	Gram
s	Second

d	Thickness
eV	Electron Voltage
M	Molarity
h	Planck constant
ν	Frequency
I_{ph}	Photocurrent
c	Speed of light
q	Charge of electron
τ_f	Fall time
τ_r	Raise time
E_g	Energy gap
R°	Free radical
V	Volume
T	Transmission

**PEMBENTUKAN DAN PENCIRIAN FILEM DARIPADA
PENGEKSTRAKAN TOMATO UNTUK PENGESAN FOTO UV**

ABSTRAK

Konsep baru pengesanan foto UV dengan ciri-ciri kuasa dan biodegradable diperlukan untuk menyelesaikan masalah tenaga dan alam sekitar. Dalam kerja ini, filem nipis tomato digunakan sebagai lapisan organik aktif dalam pengesanan foto UV kerana tomato kaya dengan antioksidan. Apabila sinaran UV berinteraksi dengan antioksidan, pemindahan elektron yang melibatkan pelepasan dan perangkapan elektron mungkin berlaku. Fenomena pengangkutan elektron ini boleh dimodulasi dengan mengaplikasi sumber kuasa luaran. Dengan konsep ini, tomato telah digunakan untuk penyelidikan ini. Kesan suhu pengeringan (60°C, 80°C, 100°C, 120°C dan 140°C) pada sifat struktur, kimia, optik, elektrik dan pengesan filem nipis tomato telah disiasat. Pengesanan foto UV terdiri daripada struktur berlapis substrat kaca / lapisan aktif filem nipis tomato / elektrod aluminium interdigitated. Suhu pengeringan optimum peranti adalah 120 °C, perbezaan peratusan responsif tertinggi (70.6%) di rantau positif direkodkan dan diukur pada 0 V dengan kecekapan kuantum (η) iaitu $2.53 \times 10^{-7}\%$, R iaitu 0.0519×10^{-6} A / W, dan D^* iaitu 0.7645×10^{21} Jones. Tanpa bekalan kuasa luaran (0 V), pengesanan foto berfungsi dengan responsif yang paling tinggi, menjadikannya sebagai penjimatan tenaga dan peranti berkuasa diri. Kemudian, ia diikuti dengan membandingkan sifat-sifat tomato filem nipis yang dihasilkan daripada ekstrak tomato dengan dan tanpa asid oksalik. Peranti yang diekstrak dengan menambahkan dengan dan tanpa asid oksalik pada 120°C, perbezaan peratusan responsif tertinggi (89.60%) di kawasan positif dicatatkan dalam pengesanan foto dengan filem nipis tomato dan asid oksalik dikeringkan pada 120°C dan diukur pada 5 V dengan kecekapan kuantum (η) iaitu $1.0335 \times 10^{-5}\%$, R

iaitu 2.1178×10^{-6} A / W, dan D^* iaitu 1.1649×10^{21} Jones. Ini menunjukkan bahawa filem nipis tomato kering pada suhu tertentu ini adalah yang terbaik digunakan untuk mengesan UV-C (254 nm) pada 0 V dan UV- (302 nm) pada 5 V, dan nilai masing-masing adalah $2.53 \times 10^{-7}\%$ dan $7.7384 \times 10^{-6}\%$. Masa tindak balas untuk menaikkan dan menurunkan semua ujian kurang daripada 0.3 s yang diperhatikan dalam kerja ini membolehkan filem nipis tomato digunakan sebagai pengesanan foto UV pada gelombang spesifik ini. Di samping itu, prestasi peranti bergantung kepada kehadiran kumpulan berfungsi, kekasaran permukaan dan indeks biasan filem nipis tomato. Parameter ini telah menjejaskan penyerapan UV yang secara langsung mempengaruhi pembentukan radikal dan aktiviti pemotongan dalam tomato, yang membawa kepada tindak balas semasa peranti. Oleh itu, tindak balas yang cepat, responsif yang tinggi, pengesanan dan kestabilan yang tinggi membolehkan peranti filem nipis tomato yang diperolehi digunakan sebagai pengesanan foto UV.

FORMATION AND CHARACTERIZATION OF THIN FILM FROM THE EXTRACTION OF TOMATO FOR UV PHOTODETECTOR

ABSTRACT

New concept of UV photodetector with self-powered and biodegradable features are needed to solve energy and environment. In this work, thin-film tomato was used as an active organic layer in UV photodetector because tomato was also rich in antioxidant. When UV radiation interacts with antioxidant, transfer electrons that involve either releases and traps of electrons may occur. This electron transportation phenomenon can be modulated by applying an external power source. With this concept, tomato has been used for this research. In addition, the effect of drying temperature (60°C, 80°C, 100°C, 120°C and 140°C) on the structural, chemical, optical, electrical and sensing properties of thin-film tomato was studied. The UV photodetector was made of a sandwich structure of glass substrate/thin-film tomato active layer/ interdigitated aluminium electrode. The optimum drying temperature of the device was 120°C, the highest responsivity percentage difference (70.6%) in positive region is recorded and measured at 0 V with quantum efficiency (η) of 2.53×10^{-7} %, R of 0.0519×10^{-6} A/W, and D^* of 0.7645×10^{21} Jones. Without external power supply (0 V), and yet the photodetector works with the highest responsivity, makes it as an energy saving and self-powered device. Then, it is followed by comparing the properties of thin-film tomato produced from tomato extract with and without oxalic acid. The fabricated using tomato extracted by adding with and without oxalic acid at 120°C, the highest responsivity percentage difference (89.60%) in positive region is recorded in photodetector with thin-film tomato with oxalic acid dried at 120°C and measured at 5 V with quantum efficiency (η) of 1.0335×10^{-5} % , R of 2.1178×10^{-6} A/W, and D^* of 1.1649×10^{21} Jones. Above

these results indicate that the thin-film tomato dried at this specific temperature is the best to be used to detect UV-C (254 nm) at 0 V and UV- (302 nm) at 5 V, respectively, and their respective η values are 2.53×10^{-7} % and 7.7384×10^{-6} %. The response time for raising and falling for all testing less than 0.3 s observed in this work enables the thin-film tomato being used as a UV photodetector at this specific wavelength. In addition, the performance of the device dependent on the presence of functional groups, the surface roughness and refractive index of thin film tomato. These results had affect the UV absorption that directly influence the radical formation and scavenging activity in tomato, leading to current response of the device. Therefore, the quick response, high responsivity, high detectivity and stability enable the obtained thin-film tomato device to be used as UV photodetector.

CHAPTER ONE

INTRODUCTION

1.1 Research Background

The Sun is the most powerful ultraviolet (UV) source, and living species of the Earth's ecosystem are affected by the solar UV radiation, which is usually classified into three bands, UV-A (400–320 nm), UV-B (320–280 nm) and UV-C (below 280 nm) as shown in Figure 1.1. UV-A is a long-range UV radiation between 320 and 400 nm. Although UV-A is not as energetic as the other two types of UV, UV-A can penetrate deep into our skin (dermis). This can cause immediate tanning and premature skin aging, over certain period of time, it may cause skin cancer (Wang et al., 2014). UVA is not readily absorbed by the ozone layer, about 95% gets through, and it usually involves in stimulating photosynthesis and synthesizing vitamins and biochemical compounds (Monory, et al., 2003) and it is an important radiation in maintaining the quality of food supply for mankind (Ahmed and Robinson, 1998).

UV-B is a short-wave UV radiation between 280 and 320 nm. A large amount of UV-B is absorbed by the ozone layer, only 5% reaches planet's surface. It has deteriorating effect on organic materials and affecting output of crops (Pancotto et al., 2003, Chen et al., 2015, Corbineau et al., 1995, Predieri et al., 1995, Soheila, 2000, Nara and Takeuchi, 2002, Chang et al., 2003). UV-B (280-320) is a radiation source for various applications, namely in medical imaging (Grundfest, 1999), forensic analysis (Smith and Lam, 2010), protein analysis and DNA sequencing (Karczemska and Sokolowska, 2002).

UV-C, with wavelengths between 100 and 280 nm, is very energetic. It is very dangerous to all forms of life, even with short exposure. However, UV-C radiation has been filtered out by ozone layer and it never reach the earth. This type of UV is technological useful whereby it can be engineered to kill bacteria and germs (Bolton and Cotton, 2008, Kowalski, 2009, Rauth, 1965, Monroy et al., 2003), disinfection and decontamination (Knight, 2004), forensic analysis (Smith and Lam, 2010), protein analysis and DNA sequencing (Karczemska and Sokolowska, 2002), space observation (VUV/EUV ranges) (Malinowski et al., 2010), and DUV(193 nm)/EUV(13.5 nm) lithography (Association, 2001) and flame detection (Monroy et al., 2003).

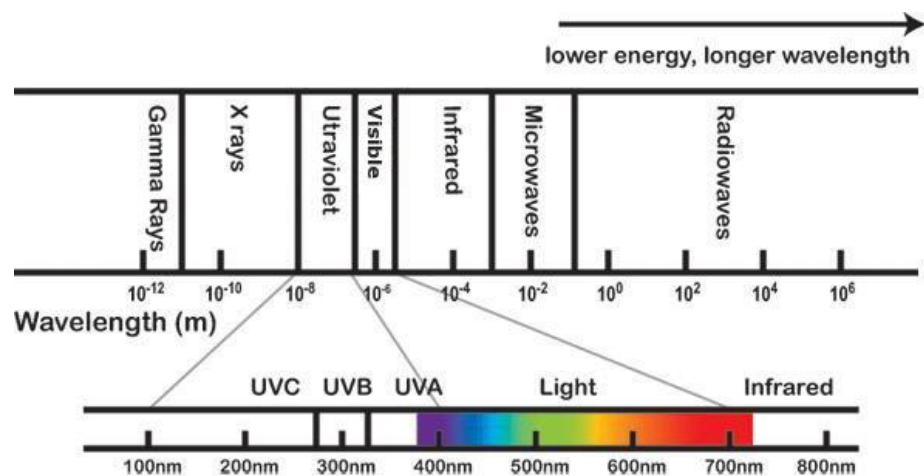


Figure. 1.1: Electromagnetic radiation spectrum (Kerker, 2016).

UV photodetectors have been used in various areas, whereby its application including biological and chemical sensors (detecting ozone, pollution level, organic compound and biological agents), flame detection (fire alarm, missile warning or combustion engine control), spatial optical communications (intra-and inter-satellite secured communications), emitter calibration (UV dosimetry and UV lithography) and

astronomical studies (Omnes, 2010, Omnès et al., 2007, Monroy et al., 2003). Among these area, the flame detection, biological and chemical sensor are related to this work because of they are working in the UV-B and UV-C range (Monroy et al., 2003, Predieri et al., 1995). For practical applications, the photodetector must be flexible, intelligent and multifunctional in design. Currently, the most widely used semiconductor-based photodetector materials is silicon (Si) because of the UV responsivity of Si-based UV photodetector are ranging from 100-350 nm (Shi and Nihtianov, 2012, Hwang et al., 2016, Kim et al., 2016, Hwang et al., 2015, Solt et al., 1996, Scholze et al., 2006, Scholze et al., 2002). Wide and direct bandgap semiconductors such as SiC, III-nitrides including AlN, GaN, InN and selected II-V compounds that have been used as the sensing materials for UV detection as they are having better spectral selectivity if compared with Si-based photodetector (Consonni and Feuillet, 2014, Leung et al., 2010, Kim et al., 2015, Zhou et al., 2016, Parida et al., 2017).

Regardless whether the sensing materials are made of Si or those typical wide bandgap semiconductors, they are all sharing a common attribute that is inability to degrade in a short period of time after lifetime of the photodetector has expired. Therefore, disposable of the photodetector may create a huge environmental issue (Alaie et al., 2015, Yang et al., 2017, Moore, 2007). As a result, intensive effort has been initiated to search for materials that can easily degrade with least toxicity and ease of fabrication aiming to reduce electronic waste (Perkins et al., 2014, Hong et al., 2015). One of the solutions is to employ natural organic materials to fabricate photodetectors and other electronic devices that can be used for a designated period of time with

acceptable performance. There are no reported work of using natural organic materials for UV photodetector application. However, there are some works using natural organic materials to produce electronic devices such as memory, battery, transistor, etc (Kim et al., 2009, Eder et al., 2004, Fu et al., 2017). The materials are carotenoid (Głowacki et al., 2011), β -carotene (Shah and Brown, 2005), aloe vera (Lim and Cheong, 2015, Lim et al., 2017), silk (Kim et al., 2009), leather (Martins et al., 2009) have been used as an active or passive material for the fabrication of biodegradability and environmental sustainable devices.

In this work, tomato as a natural source of organic material has been chosen to be used as an active source to detect UV radiation. Tomato is a member of Solanaceae family and it is considered as a kind of “Mediterranean diet”, which is strongly associated with the ability to reduce chronic degenerative diseases (Agarwal and Rao, 2000, Rao et al., 1998, Bulling, 2013, Rao and Agarwal, 1999) due to its various pharmacologic properties (Giovannucci et al., 2002, GIULIANO, Hartje et al., 2000, Hamid et al., 2010). In addition, tomato is also rich in antioxidant (Clinton, 1998, Li et al., 2011d, Elbadrawy and Sello, 2016, Kotíková et al., 2011). Various vitamins available in tomato are believed to be the source of antioxidant. When UV radiation interacts with antioxidant, transfer electrons that involve either releases and traps of electrons may occur. This electron transportation phenomenon can be modulated by applying an external power source. With this concept, tomato has been used for this research. In order to use it as a solid-state UV photodetector, the extracted tomato juice must be formulated, deposited on a glass substrate, and processed it into a thin film. This natural material has yet been employed for this kind of work. Similarly, there are no

related works that used natural organic materials for this purpose. Majority of the works utilized synthetic polymers (Zhang et al., 2009, Persano et al., 2015, Xu et al., 2010, Burkhardt et al., 2012, Liao et al., 2012, Shao et al., 2013b) or inorganic materials (Koide, 2008, Jain et al., 2000, Auret et al., 2001, Young et al., 2006, Xu et al., 2006). The only available applications of tomato in electronic devices are for electronic nose tongue (Gomez et al., 2008) based on tomato juice but not in the form of thin film. Without a solid form of geometry, it is impossible to be used as a building block for a solid-state device application. Therefore, it is extremely important to process the tomato juice into a solid thin film. However, up-to-date, there is no literature related neither to the processing condition nor the property of thin-film tomato. Therefore, in this work, systematic investigation has been performed in order to extract, formulate, and process the extracted tomato juice into a functional solid thin film with the objective of using this active layer to detect selective UV radiation with minimum operating power that is able to address issue of electronic waste.

1.2 Problem statements

Incorporation of the UV photodetector in flexible, intelligent and wearable design for the application of health and safety has been an intensive research area (Alaie et al., 2015, Yang et al., 2017). Not limited to this, the photodetectors must be able to be disposed safely after their lifetime in order to minimize impact to the environment. Researches on human and environmental-friendly materials have been the focus to address this issue (Perkins et al., 2014, Yang et al., 2017). However, there is no reported work that uses natural organic materials forming into a thin film and use for UV detection purposes. Therefore, in this research, tomato has been selected as a choice to