NATURAL DYE FOR DYE SENSITIZED SOLAR CELLS (DSSCs) USING MANGOSTEEN PERICARP AND ADENIUM OBESUM

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CHEMICAL ENGINEERING
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Dissertation submitted in partial fulfillment of
the requirements for
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Perak

CERTIFICATION OF APPROVAL

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Approved by,

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(Dr. Wan Zaireen Bt Nisa Yahya)

UNIVERSITI TEKNOLOGI PETRONAS
BANDAR SERI ISKANDAR, PERAK
September 2015

CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own concept as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources of persons.

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CHANG OON HANG

ABSTRACT

Dye sensitized solar cells is now a significant research in the last two decades due to its fundamental solution in the energy conversion at lowest cost. Besides that, the ease of fabrication, flexibility and widely available materials have made the dye sensitized solar cell a promising technology towards future innovation in low cost solar cell. Although the novel organic synthetic dyes provide a high efficiency and durability compared to natural dye, there are several limitations such as high cost and complexity of synthesis and toxicity of materials. These setbacks had opened up for further research of utilizing the natural sensitizers which is environmental friendly and ease in preparation. The main objective of this research is to determine the optimum condition of types of solvent, extraction temperature and pH value for extraction in improving the efficiency of natural dye using mangosteen pericarp and adenium obesum flower. The mangosteen pericarp extracted with acetone solvent shows the highest light absorption intensity and power conversion efficiency value. The optimum pH condition for the mangosteen pericarp extract is at pH 1.93 with the highest efficiency. For the adenium obesum extract, the optimum temperature for the extraction is at 40ºC and the adenium obesum extracted at pH 4.01 shows a high efficiency. The mixture of mangosteen pericarp-adenium obesum shows significantly the highest light absorption intensity and photoelectrochemical values compared to its pure extracts. The optimum pH for the mixture extract is at pH value of 2.93.

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LIST OF ABBREVIATION

CdTe	Cadmium Telluride
DSSC	Dye Sensitized Solar Cell
I_{sc}	Short Circuit Current
FF	Fill Factor
PCE	Power Conversion Efficiency
Pt	Platinum
V_{oc}	Open Circuit Voltage

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CHAPTER 1

INTRODUCTION

1.1 Background Study

The concept of harnessing the Sun energy has raised interest to scientist and extensive research has been executed in order to figure out and copy the photosynthesis process which uses the sunlight to convert water to its consecutive components and carbon dioxide into carbohydrates (Kalyanasundaram, 2010). With its extensive study, the theory to substitute the fossil fuels with the solar power radiation from the Sun was proposed in 1912 (Ciamician, 1912).

Photovoltaic device converts sunlight into electricity without the effect of pollution of the environment and sound to the surroundings which enables it to be a renewable and reliable source (Ludin, Ludin, Al-Alwani Mahmoud, Bakar Mohamad, & Kadhum). The mechanism of photovoltaic technology uses the generation of charge between the interface of two different conduction materials (Jasim, 2011). The first panchromatic film which renders the realistic images into black and white colours with the discovery of the method to intensify the photographic emulsion sensitivity using silver halide dye to create black and white photographic films. Silver halide chemicals are insensitive to the visible light due to its low band gaps at 2.7–3.2 eV. Hence, this discovery has led to the first significant study on the dye sensitization of semiconductors in the photovoltaic technology (Ludin et al., 2014).

The conventional photovoltaic device remains to be the first generation solar cell made of pure silicon material. Although the production cost for the silicon based solar cell are quite low, the cost still relies heavily on the price and availability of the main raw material, silicon. Thus, the emergence of the second generation thin film photovoltaic technology based on the amorphous silicon (Wronski, 2000) and Cadmium Telluride competes with the conventional silicon based solar cell and able to double its industry production capacity with its simplicity of manufacturing and a wider application range. However, there were several handicaps to the thin film technologies such as the usage of toxic raw materials and efficiency heavy reliance on the temperature. The first and second generation solar cell are based on single junction which has a limited efficiency. The third generation solar cell overcomes the Shockley and Queisser limit which is the maximum conversion and aims to have large scale generation of electricity at a competitive low price compared to the first and second generation solar cell in order for the photovoltaic technology to be the most economical energy production in the future. The dye sensitized solar cell technology is between the second and third generation solar cell with a low production cost and high material availability (Pettersson, Hagfeldt, Boschloo, & Kloo, 2010).

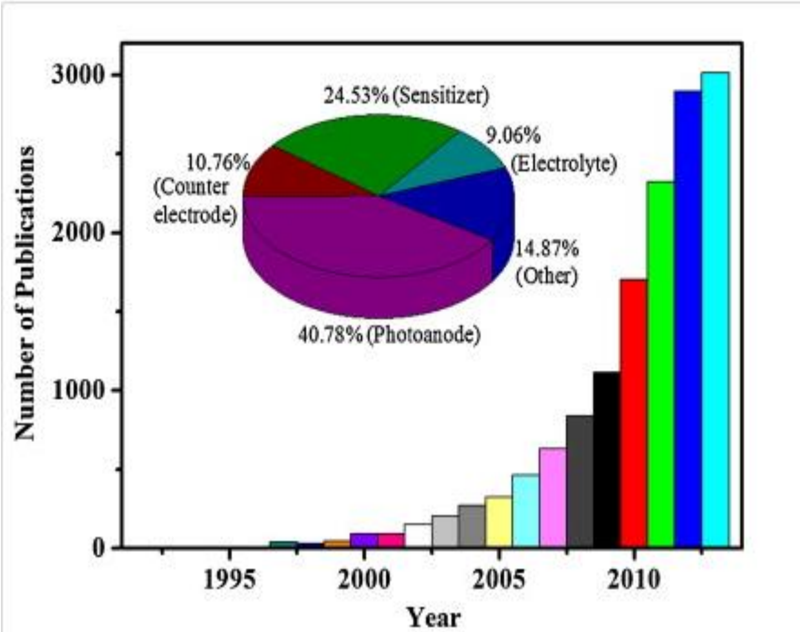

Figure 1.1 Evolution of the number of publications for ‘sensitized solar cells’

As seen from Figure 1.1 taken from ISI Web of Science by Thompson Reuters, dye sensitized solar cell has been researched extensively on all the components parts of the cell especially the photoanode component and attracted the research world as well as the industrial world. The dye sensitized solar cell technology has established several research groups around the world especially in Europe, Japan, Korea, Australia and China. Besides that, several multinational companies such as BASF, Toyota, Sharp, Sony and Panasonic also showed interest in the dye sensitized solar cell technology with several research companies such as Dyesol were selling the raw materials and components of dye sensitized solar cell (Pettersson et al., 2010) .

1.2 Problem Statement

The Nobel laureate Richard Smalley stated that one of the major problems faced by mankind is energy and environment issues. The increasing population corresponds with the rising of energy consumption which accelerates the depletion of oil reserves and high level of pollution and greenhouse effect caused by several factors such as the combustion of fossil fuels. Thus, several research has been carried out in order to find a suitable replacement by using the renewable and green energy with low cost of fabrication as well (Gong, Liang, & Sumathy, 2012). Although the shale gas technology has emerged to be the promising resource of energy with its abundance and cheap cost in replacing the depleting fossil fuel, the solar cell can be an alternative towards further applications of generating electricity at a global scale with minimal damage to the environment as well as flexibility in design.

Harnessing the clean and cheap energy source of the Sun is a promising technology in overcoming the hurdle of environmental issues of global warming and greenhouse effect. Photovoltaic devices is based on the technology of converting solar energy to electrical energy (Nazeeruddin, Baranoff, & Grätzel, 2011) and considered as the most efficient technology compared to other sustainable energy sources. At the starting of the solid state dye sensitized solar cell, inorganic dye such as the ruthenium complexes exhibit best efficiency and become the ideal sensitizer. Due to its limited availability of raw materials and costly price as well as the difficulty in synthesizing the ruthenium complexes, scientist has been looking for alternatives to replace it with a cheaper alternative of synthesizing synthetic organic dye for the DSSC. Although some of the synthetic organic dye with the structure of $D\text{-}(\pi\text{-}A)^2$ exhibits high efficiency compared to natural dye, the organic dyes chemically synthesized require complex steps with expensive reagents and catalyst needed and might produce harmful by products from the reactions which might be toxic.

Hence, in order to achieve a lower synthesizing cost, natural dyes are able to answer up to the challenge as it is abundantly available and requires simple preparation method only. This can reduce the cost for synthesizing the dyes and natural dyes do not have toxic properties which enables it to be an environmental friendly alternative. Several researchers had extracted natural dyes from the flowers, fruits, leaves, and others but obtain very low efficiency state (Shalini, Balasundara prabhu, Prasanna, Mallick, & Senthilarasu, 2015). Hence, it is critical to ensure that the extraction condition is at its optimum level to increase the efficiency of natural dye as well as the stability of the dye to withstand extensive exposure to sunlight.

1.3 Objectives

The main objective of this research is to determine the optimum extraction condition for the natural dye of mangosteen pericarp and adenium obesum flower. The pH level, extracting temperature and the effect of type of extracting solvent used in the extraction of natural dye are extremely crucial to improve the photovoltaic performance of the natural dye of mangosteen pericarp and adenium obesum flower.

To achieve the main objective, the sub objectives of the research as listed below:

a) To know how does the various type of solvent such as water, ethanol, chloroform and acetone used for the extraction of mangosteen pericarp affects the photovoltaic performance and efficiency of DSSC.

b) To examine the effect of pH level of the extracted dye from mangosteen pericarp and adenium obesum flower on the efficiency level and the stability of natural dye.

c) To determine the optimum extracting temperature for adenium obesum flower for maximum absorption spectrum wavelength for absorption into the titanium oxide and photovoltaic performance.

d) To compare the absorption spectrum, efficiency and photovoltaic performance of DSSC between natural dyes extracted from mangosteen pericarp and adenium obesum flower.

1.4 Scope of Study

Since there is abundance of the natural resources of plants, leaves and others in Malaysia due to its tropical weather and rainforest, the scope of the research will be limited to using mangosteen pericarp as shown in Figure 1.2 and adenium obesum flower as shown in Figure 1.3 as the natural dye source. The mangosteen pericarp is suitable due to its complex molecular structure, xanthone of hydroxyl that enables a covalent bonding between the titanium dioxide and mangosteen pericarp as shown in Figure 1.4. The adenium obesum flower structure is not researched and being used for the extraction for dye in dye sensitized solar cell.

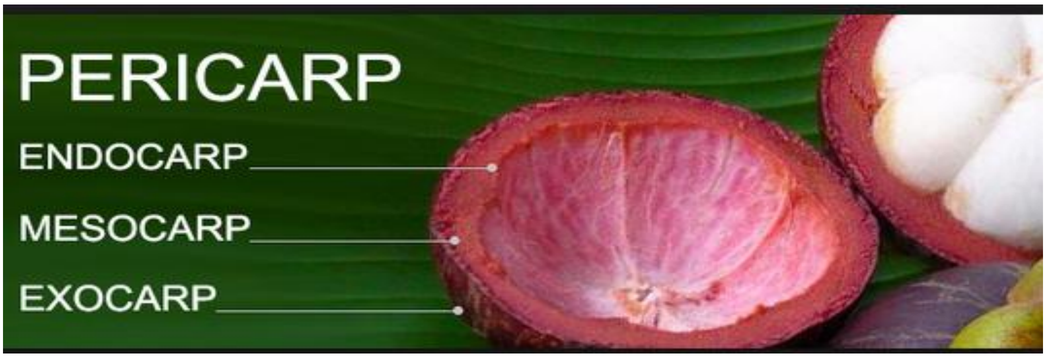

Figure 1.2 Mangosteen Pericarp and its section

Figure 1. 3 Adenium Obesum Flower

The mangosteen pericarp and adenium flower are abundant in Malaysia and a suitable representatives for the comparison of fruits and flower extracted natural dye. Thus, the project is relevant in optimizing the extraction condition for highest efficiency for mangosteen pericarp and adenium obesum as well as feasible within the time frame with its easy preparation.

CHAPTER 2

LITERATURE REVIEW

2.1 Dye Sensitized Solar Cell

Photovoltaic devices use the charge separation at an interface of two materials of different conductivity in order to produce electricity. The devices are usually solid-state junction devices that are made of silicon in the semiconductor industry. Then, the third generation of photovoltaic cells based on the nanocrystalline and conducting polymer films challenges the inorganic conventional photovoltaic device with its low cost of fabrication by replacing the system with electrolyte, liquid, or gel in the photoelectrochemical cell. The dye sensitized solar cells are based on the concept of photoelectrochemical cell with the optical light absorption by the sensitizer dye and a wide band gap semiconductor of nanocrystalline morphology. The device exhibits a power conversion efficiency of 12 % in diffuse daylight and high stability researched by Gratzel.(O'Regan & Grätzel, 1991).

There are several types of photovoltaic devices such as the organic photovoltaic device and dye sensitized solar cell. The typical type of conventional organic photovoltaic device uses organic materials with the donor and acceptor electrons to form a heterojunction that separates the exciton into two carriers. Then, the carriers transfers to the electrode with the same organic materials which generates the exciton. This photovoltaic device faces the difficulty to achieve a good light harvesting properties and carrier transporting attributes. Thus, the dye sensitized solar cells (DSSC) separate the two specification to have a charge generation at the dye and charge transfer at the electrolyte. The spectral attribute improvement can be executed by modifying the structure of dye, optimizing the semiconductor and electrolyte properties (Nazeeruddin et al., 2011).

2.1.1 Working Operations of Dye Sensitized Solar Cells

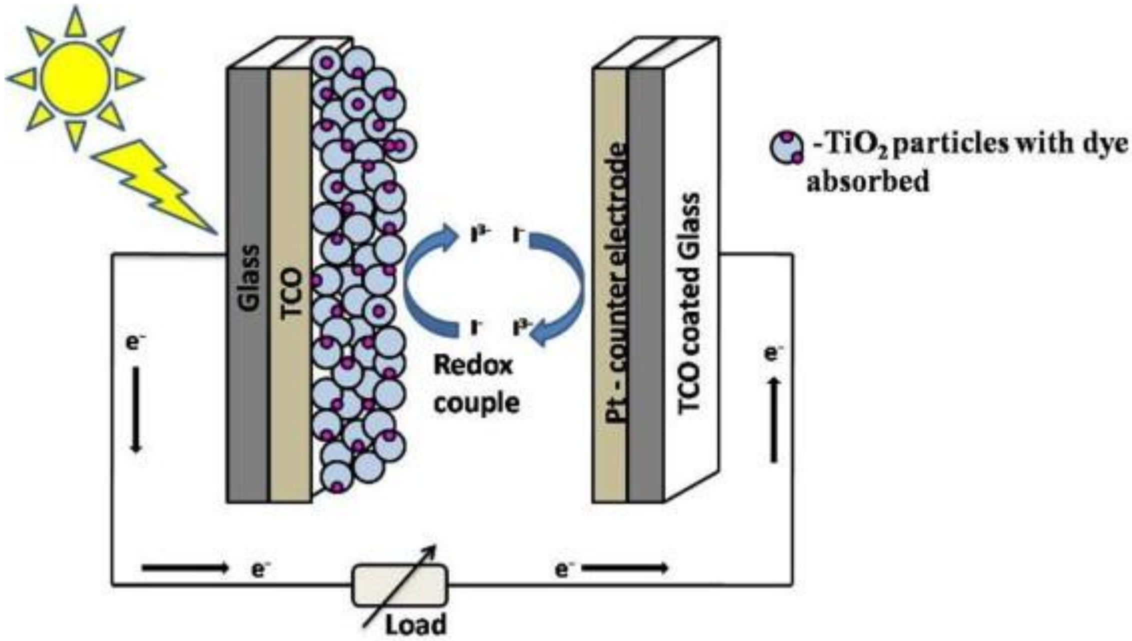

Figure 2.1 Schematic Structure of DSSC(Shalini et al., 2015)

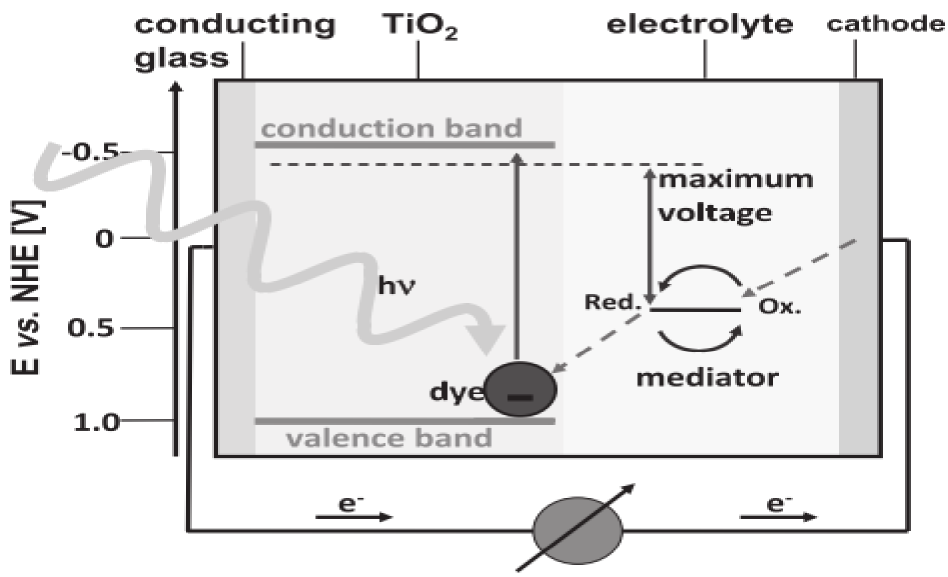

Figure 2.2 Operating Principles and Energy Level Diagram of DSSC(Andrade, Ribeiro, & Mendes, 2011).

- As shown above in Figure 2.1 above, the schematic representation of the dye sensitized solar cell. The dye sensitized solar cell converts solar radiation into electric current by undergoing the “artificial photosynthesis”(Kalyanasundaram, 2010).

The charge separation process in DSSC consists of the few steps which repeats to form a cycle as indicated in Figure 2.2. The steps are as listed below (Andrade et al., 2011).

(i) The photoanode absorbs the incident solar energy and causes the electrons in the dye sensitizer to be excited from the ground state to the excited state as indicated in equation (1).

$$S_{(adsorbed)} + hv \rightarrow S^{*}_{(adsorbed)} \quad (1)$$

(ii) The electrons from the excited state are injected into the conduction band of semiconductor which allow the dye sensitizer to be in an oxidized state as shown in equation (2).

$$S^{*}_{(adsorbed)} \rightarrow S^{+}_{(adsorbed)} + e^{-}_{(injected)} \quad (2)$$

(iii) The electrolyte donates the electron to restore the dye to its initial neutral state as seen in equation (3). At the same time, the electron travels the external semiconductor circuit and reaches the counter electrode and reduce the redox mediator though the equation (4). This completes the cycle.

$$S^{+}_{(adsorbed)} + \frac{3}{2}I^{-} \rightarrow S_{(adsorbed)} + \frac{1}{2}I_3^{-} \quad (3)$$

$$I_3^{-} + 2e^{-}_{(cathode)} \rightarrow 3I^{-}_{(cathode)} \quad (4)$$

However, in addition to forwarding transfer process shown above, backward charge transfer process can occur and reduces the efficiency level of the dye sensitized solar cell. The backward charge completes the cycle with the recombination of the injected electrons either with oxidized sensitizer indicated in equation (5) or with the oxidized redox couple at the TiO_2 surface in equation (6) (Nazeeruddin et al., 2011).

$$S^{+}_{(adsorbed)} + e^{-}_{(TiO_2)} \rightarrow S_{(adsorbed)} \quad (5)$$

$$I_3^{-} + 2e^{-}_{(TiO_2)} \rightarrow 3I^{-}_{(anode)} \quad (6)$$

Several requirements are required in order to reduce the effects of backward transfer charge process such as ensuring that the charge transfer to the semiconductor occurs within the quantum yield (Grätzel, 2004), the rate of electron injection must be above the rate of electrons decay from its excited state to neutral state (Anderson & Lian, 2005), the LUMO level of the sensitizer must be more negative than the conduction band and HOMO level must be more positive than the redox potential (Hara et al., 2003).

2.1.2 Components of Dye Sensitized Solar Cell

The photosensitizer, photoanode, electrolyte and counter electrode are the four major components of dye sensitized solar cell (Ye et al., 2015).

Multiple alternatives and approaches to alter the photosensitizer has been done to improve the efficiency level. There are fulfillments of the attributes of an effective photosensitizer such as high intensity of light absorption in the visible region, high injection of electron into the conduction band of semiconductor and functional groups that are able to attach to the titanium oxide (Smestad, 1998). The efficiency level of the dye sensitized solar cell is mostly dependent on the molecular structure of the sensitizers. There are three classes of photosensitizers developed currently: the metal complex sensitizers, metal-free organic sensitizers, and natural sensitizers (Narayan, 2012). Several attempts of synthesizing metal-free organic sensitizers are executed but could not rival the efficiency of transition metals based photosensitizers.

Nanostructured semiconductor films are the main framework of DSSC novel photoanodes. The photoanode serves dual functions as the support for sensitizer bonding and transporter of photo-excited electrons from sensitizer to external circuit. Hence, a large surface area of photoanode is necessary to ensure high dye loading and high DSSC efficiency. Moreover, a fast charge transport rate is required to ensure high electron collection efficiency as well. These two properties are the crucial defining characteristics of an ideal photoanode (Maçaira, Andrade, & Mendes, 2013).

The electrolyte is a crucial component in the dye sensitized solar cell. The composition of electrolyte is a very crucial parameter in determining the performance of dye sensitized solar cell. The requirements of an electrolyte is chemically inert, optically stable and able to withstand high heat to ensure the degradation of the dye does not occur. Some of the electrolytes used in the dye sensitized solar cell are divided according to its characteristics such as liquid electrolyte, polymer electrolyte (Su'ait, Rahman, & Ahmad, 2015), quasi-solid electrolyte and solid-state hole transport conductor (Ye et al., 2015).

The counter electrode of DSSC functions to transferring the elections from the circuit to the electrolyte. Hence, the counter electrode must have a high charge transfer resistance and a high exchange current densities (Anandan, 2007). Platinum has been the ideal and commercially used material for the counter electrode due to its ability as the catalyst for reduction of electrolyte (Luque & Hegedus, 2011).

2.1.3 Developments of Novel Photosensitizers for DSSC

The efficiency of the DSSC is significantly affected by the usage of a good photosensitizer. Lots of efforts have been done for developing and synthesizing dyes whose absorption can be extended in the near infra-red (NIR) region. Metallic dyes are more stable and efficient, thus frequently researched out previously. Various modification on metallic dyes have also been worked out in the structure by complexing it with different groups. Metallic dyes are also better in its higher absorption properties(Bose, Soni, & Genwa).By far the most important sensitizer showing highest conversion efficiency and long term stability are that of Ruthenium and Osmium (Lan, Wu, Lin, & Miaoliang, 2012). Some of the important molecular structures of three important Ru-dyes are shown in Figure 2.3.

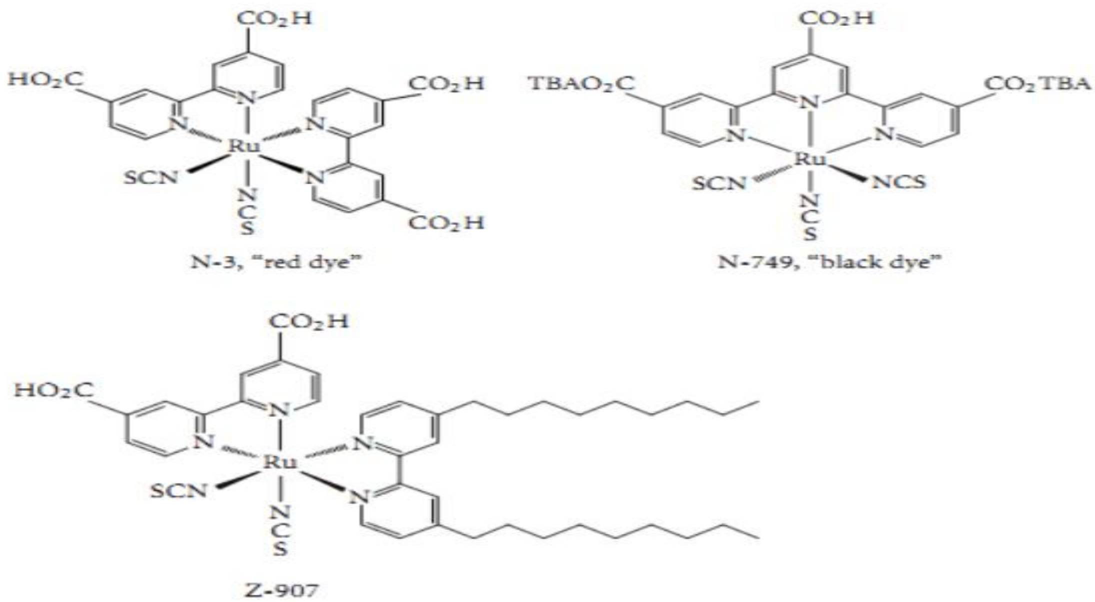

Figure 2. 3 Molecular Structure of Ruthenium Complex(Bose et al.)

Several attempts have been made to synthesize novel dyes that could improve the overall efficiency of the DSSC. The sensitizers based on noble metals such as ruthenium, osmium, and rhenium showed high efficiency but their cost and complicated synthesis process and toxicity restrict their industrial applications. In order to reduce the cost of fabrication of DSSCs, researchers have turned their focus in organic sensitizers as replacement for metal sensitizers. The organic dyes offers several possibilities and flexibility by improving its molecular structure in order to bring about improvement in the light harvesting ability of dyes as well as absorption into the photoanode. Thus, optimal DSSCs have been fabricated with several novel organic dye developed over the recent years. Table 2.1 indicates the types of novel organic dye developed over the recent years. Based on the table 1, the highest efficiency level is the D-A-pi-A indoline dyes of XS45 type which showed and efficiency of 6.9 %. However, the synthesizing cost remains expensive and thus restricting in applying towards commercial applications. In order to reduce the cost of DSSCs further, researchers are also focusing on natural sensitizers such as betacyanin and anthocyanin or chlorophyll extracted from natural resources of plants, fruits, and others . (Chawla & Tripathi, 2015).

Table 2.1: Photovoltaic parameters of different dyes fabricated(Sugathan, John, & Sudhakar, 2015)

Sensitizer No.	Novel Dyes	J_{sc}(mA /cm)	V_{oc}(V)	FF	Efficiency (%)	Reference Cite

1	Artificial chlorin-type sensitizers	Type					(Liu et al., 2013)
		Dye-1	8.55	0.61	0.69	3.65	
		Dye-2	7.21	0.60	0.67	2.92	
		Dye-3	4.35	0.59	0.72	1.85	
		Dye-4	8.37	0.60	0.71	3.60	
2	D-A-pi-A indoline dyes	Type					(Wang et al., 2014)
		XS45	14.8	0.68	0.68	6.90	
		XS46	13.3	0.64	0.69	5.87	
3	BODIPY series of dyes derived from C219	Type					(Mao et al., 2014)
		UY1	6.17	0.49	0.71	2.14	
		UY2	6.46	0.50	0.71	2.28	
		UY3	9.44	0.55	0.63	3.24	
		UY4	12.16	0.59	0.68	4.89	
4	Novel organic dyes	Type					(Wu et al., 2013)
		J5	10.01	0.41	0.64	2.6	
		J6	9.48	0.33	0.64	2.0	
5	Cyclometalated ruthenium sensitizers	Type					(Li et al., 2014)
		NC102	8.15	0.63	0.71	3.64	
		NC103	9.45	0.63	0.71	4.22	
6	New organic dyes	Type					(Reddy et al., 2014)
		CSOR G6	9.19	0.64	0.72	4.30	
		CSOR G7	11.56	0.72	0.71	6.00	
		CSOR G8	10.84	0.69	0.72	5.40	
		CSOR G9	12.07	0.72	0.68	6.00	
7	New dyes for P-type DSSC	Type					(Zhu, Yang, Zhong, & Li, 2014)
		T3	4.01	0.14	0.33	0.19	
		T4	1.69	0.12	0.29	0.06	

2.1.4 Natural Dyes Sensitizers for Dye Sensitized Solar Cells

The natural dyes had been extensively used in dye sensitized solar cell due to its low fabrication cost, simple extraction, environmentally friendly , and non-toxicity (Dai & Rabani, 2002).

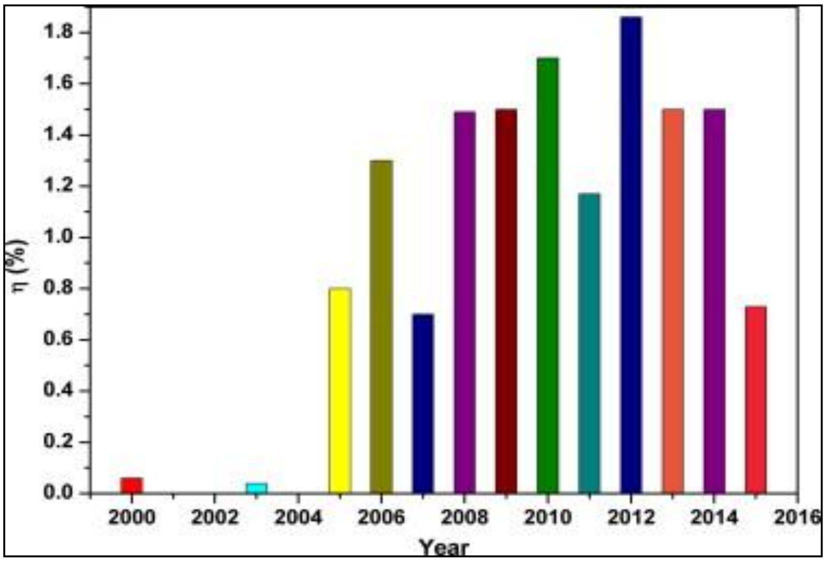

Figure 2.4 Plot of progress in efficiency (η) reported for DSSC utilizing different types of natural sensitizers during the last 5 years.(Shalini et al., 2015)

Based on Figure 2.4, a highest efficiency of 1.86 % is achieved in the year 2012 for punicagrantum peel (Hernández-Martínez, Estevez, Vargas, Quintanilla, & Rodriguez, 2012). Natural dye of fruits, flowers, leaves and bacteria exhibit various colours and contain several pigments that can be extracted and utilized as dye in DSSC (Ho Chang & Lo, 2010). The advantages of using natural dyes as photosensitizers in DSSC are due to their large absorption coefficients in the visible region (Luo et al., 2009).

Plant pigments exhibit electronic structure that interact with the sunlight and change the wavelengths that are either transmitted or reflected by the plant tissue. This process causes the occurrence of plant pigmentation and each pigment is described from the respective wavelength of maximum absorbance (λ_{max}) and the colour perceived by humans (Obón & Rivera, 2006).The pigments for natural dyes are relatively easy to extract from natural products when compared to synthetic novel organic dyes. The plant pigments are categorized into chlorophyll, carotenoid, flavonoid and anthocyanin (Shalini et al., 2015). The Table 2.2 below shows the most common types of pigments found in flower and fruit colours in plants.

Table 2.2: Most common pigment types found in flower and fruit colours in plants (Obón & Rivera, 2006)

Colour	Specific pigment type	Pigment group	Examples
Cream	Flavonols or flavones	Flavonoid	Most cream flowers
Pink to red		Carotenoid	Few red flowers and fruit, e.g.*Lycopersiconesculentum*(tomato) fruit
	Pelargonidin and/or cyanidin	Flavonoid	Most pink flowers and some fruit, e.g.*Eustomagrandiflorum*(lisianthus) flowers
	Pelargonidin and/or cyanidin	Flavonoid	Most red flowers and some fruit, e.g. Malus (apple) fruit
	Anthocyanin and carotenoid mix	Flavonoid and carotenoid	A few examples, e.g. Tulipa flowers
	Betacyanin	Betalain	A few examples in the *Caryophyllales*, e.g. Bougainvillea flowers
Orange		Carotenoid	Most orange flowers and fruit, e.g.*Tageteserecta*(marigold) flowers
	Pelargonidin alone	Flavonoid	A few examples, e.g. *Pelargonium*flowers
	Anthocyanin and aurone mix	Flavonoid	Rare occurrence, e.g. *Antirrhinum majus* (snapdragon) flowers
	Anthocyanin and chalcone mix	Flavonoid	Rare occurrence, e.g. *Dianthus*(carnation) flowers
	Betacyanin	Betalain	A few examples in the *Caryophyllales*, e.g. Portulaca (purslane) flowers
Yellow		Carotenoid	Most yellow flowers and fruit
	Aurone	Flavonoid	Rare occurrence, e.g. *Antirrhinum majus* flowers
	Chalcone	Flavonoid	Rare occurrence, e.g. *Dianthus*flowers
	Betaxanthin	Betalain	A few examples in the *Caryophyllales*, e.g. Portulaca flowers
Green		Chlorophyll	All green flowers and fruit
	Delphinidin	Flavonoid	Most blue flowers and fruit
	Cyanidin	Flavonoid	Blue rare occurrence, e.g. *Ipomoea* (morning glory) flowers

Purple		Carotenoid	Rare occurrence, e.g. Capsicum (pepper) fruit
	Cyanidin and/or delphinidin	Flavonoid	Most mauve flowers, e.g. Petunia and some purple fruit, e.g. *Solanummelongena*(eggplant)
	Cyanidin and/or Delphinidin	Flavonoid and carotenoid mix	Few flowers, e.g. *Cymbidium orchids*
Black	Delphinidin	Flavonoid and carotenoid mix	Few black flowers, e.g. Viola (pansy)

Anthocyanins is the group most extensively investigated as natural sensitizers and their extracts shows maximum absorption in the range of 510 to 548 nm, depending on the fruit or solvent used (Narayan, 2012). Several researchescategorized based on flowers, fruits, leaves, seeds and others dye's performance had been investigated and listed down in Table 2.3.

Table 2.3: Photoelectrochemical parameters of natural dye for DSSC(Ludin et al., 2014)

Dye	Semiconducting oxide	λ_{max}(nm)	I_{sc}(mAcm−2)	V_{oc}(V)	FF	η (%)	Reference Cite
Flowers							
Begonia	TiO2	540	0.63	0.53	72.2	0.24	(H. Zhou, Wu, Gao, & Ma, 2011)
Rhododendron		540	1.61	0.58	60.9	0.57	
Marigold		487	0.51	0.54	83.1	0.23	
Perilla		665	1.36	0.52	69.6	0.5	
China loropetal		665	0.84	0.51	62.6	0.27	
Yellow rose		487	0.74	0.6	57.1	0.26	
Flowery knotweed		435	0.6	0.55	62.7	0.21	
Petunia		665	0.85	0.61	60.5	0.32	
Violet		546	1.02	0.49	64.5	0.33	
Chinese rose		516	0.9	0.48	61.9	0.27	
Rose		–	0.97	0.59	65.9	0.38	
Lily		–	0.51	0.49	66.7	0.17	
Hibiscus sabdariffa L.	TiO2	520	1.63	0.4	0.57	0.37	(Wongcharee, Meeyoo, &

							Chavadej, 2007)
Clitoriaternate a		580	0.37	0.37	0.33	0.05	
Erythrinavarie gata	TiO2	451,492	0.78	0.48	0.55	–	(Hao, Wu, Huang, & Lin, 2006)
Rosa xanthine		560	0.64	0.49	0.52	–	
Hibiscus surattensis	TiO2	545	5.45	0.39	0.54	1.14	(Fernando & Senadeera, 2008)
Neriumolende r		539	2.46	0.4	0.59	0.59	
Hibiscus rosasinesis		534	4.04	0.4	0.63	1.02	
Sesbaniagran diflora		544	4.4	0.41	0.57	1.02	
Ixoramacroth yrsa		537	1.31	0.4	0.57	0.3	
Red Bougainvillea glabra	TiO2	482,535	2.34	0.26	0.74	0.45	(Hernandez-Martinez, Estevez, Vargas, Quintanilla, & Rodriguez, 2011)
Violet Bougainvillea glabra		547	1.86	0.23	0.71	0.31	
Red Bougainvillea spectabilis		480	2.29	0.28	0.76	0.48	
Violet Bougainvillea spectabilis		535	1.88	0.25	0.73	0.35	
Fruits							
Tangerine peel	TiO2	446	0.74	0.59	63.1	0.28	(H. Zhou et al., 2011)
Fructuslycii		447,425	0.53	0.68	46.6	0.17	
Mangosteen pericarp		389	2.69	0.68	63.3	1.17	
Raspberries	TiO2	540	0.26	0.42	64.8	1.5	(Alhamed, Issa, & Doubal, 2012)
Grapes		560	0.09	0.34	61.1	0.38	
Citrus sinensis (Red Sicilian)	TiO2	515	3.84	0.34	0.5	–	(Calogero & Di Marco, 2008)
Solanummelongena (Eggplant)		522	3.4	0.35	0.4	–	
Cheries	TiO2	500	0.46	0.3	38.3	0.18	(Jasim, Al-Dallal, &

							Hassan, 2011)
Capsicum	TiO2	455	0.23	0.41	0.63	–	(Hao et al., 2006)
Kopsiaflavida	TiO2	550	1.2	0.52	0.62		(Nishantha, Yapa, & Perera, 2012)
Berberiesbuxifolia Lam (Calafate)	TiO2	533	6.2	0.47	0.36	–	(Polo & Iha, 2006)
Myrtuscauliflora Mart (Jaboticaba)		520	7.2	0.59	0.54	–	
Hylocereuspolyrhizus (Dragon fruit)	TiO2	535	0.2	0.22	0.3	0.22	(Ali & Nayan, 2010)
Wild Sicilian Prickly Pear	TiO2	465	8.2	0.38	0.38	1.19	(Calogero et al., 2010)
Chaste tree fruit	TiO2	548	1.06	0.39	0.48		(Garcia, Polo, & Iha, 2003)
Mulberry		543	0.86	0.42	0.43		
Cabbage-palm fruit		545	0.37	0.44	0.61		
Ivy gourd fruits	TiO2	458,480	0.24	0.64	O.49	0.09	(Shanmugam, Manoharan, Anandan, & Murugan, 2013)
Leaves							
Herbaartemisiaescopariae	TiO2	669	1.03	0.48	68.2	0.34	(H. Zhou et al., 2011)
Chinese holly		–	1.19	0.6	65.4	0.47	
Vernonia amygdalin (Bitter Leaf)	TiO2	400	0.07	0.34	0.81	0.69	(Boyo, Shitta, Oluwa, & Adeola, 2012)
spinach	TiO2	437	0.47	0.55	0.51	0.13	(H Chang et al., 2010)
Ipomoea		410	0.91	0.54	0.56	0.28	
Festucaovina	TiO2	420,660	1.18	0.54	0.69	0.46	(Hernández-Martínez et al., 2012)
Brassica olercea (Red cabbage)	TiO2	537	0.5	0.37	0.54	0.13	(Dumbrava et al., 2008)
Allium cepa (Red onion)		532	0.51	0.44	0.48	0.14	
Punicagranatum (Pomegranate)	TiO2	412,665	2.05	0.56	0.52	0.59	(Ho Chang & Lo, 2010)
Shiso	TiO2	440,600	3.56	0.55	0.51	1.01	(Kumara et al., 2006)

Jathophacurcas Linn (Botuje)	TiO2	400	0.69	0.05	0.87	0.12	(Hussain, 2013)
Lawsoniainermis (Henna)	TiO2	518	1.87	0.61	0.58	0.66	(Aduloju, Shitta, & Simiyu, 2011)
Ficusreusa	TiO2	670	7.85	0.52	0.29	1.18	(Lai, Su, Teoh, & Hon, 2008)
Rhoeospathacea		670	10.9	0.5	0.27	1.49	
Garciniasubelliptica		670	6.48	0.32	0.33	0.69	
Anethumgraveolens	TiO2	666	0.96	0.57	40	0.22	(Taya, El-Agez, El-Ghamri, & Abdel-Latif, 2013)
Parsley (Petroselinumcrispum)		666	0.53	0.44	34	0.07	
Arugula		666	0.78	0.59	42	0.2	
Seeds							
Coffee	TiO2	–	0.85	0.55	68.7	0.33	(H. Zhou et al., 2011)
Oryza sativa L. indica (Black Rice)	TiO2	560	1.14	0.55	0.52	–	(Hao et al., 2006)
Bixaarellana L. (achiote)	TiO2	474	1.1	0.57	0.59	0.37	(Gómez-Ortíz et al., 2010)
	ZnO		0.08	0.32	0.37	0.01	
Other							
Green algae	TiO2	666	0.13	0.41	21	0.01	(Taya et al., 2013)
Kelp	TiO2	670	0.43	0.44	0.62	–	(Lai et al., 2008)

2.1.5 Extraction Condition of Natural Dye for DSSC

One of the factors that might led to the low efficiency of natural dye is the extraction condition of natural dye especially temperature. There were several recent research done to improve the low efficiency of natural dye as photosensitizer by tailoring the extraction condition. The Table 2.4 summarizes the recent research executed in order to investigate the optimum extraction condition for various natural dye. From Table 4, different types of natural dye has its respective optimum extraction condition of temperature and pH value as well as exhibiting highest efficiency with different types of solvent. Hence, it is difficult to predict the optimum extraction condition for various type of natural dyes from plant and this might be due to the various concentration of pigments in the natural dye.

Table 2.4: Summary of Recent Research of Extraction Condition of Natural Dye

Natural Dye	Extraction Solvent	Research Topic	Result	Reference Cite
Red *bougain villea glabra* flower	Water	Betalain pigments as natural photosensitizers for dye-sensitized solar cells: the effect of dye pH on the photoelectric parameters	DSSCs from dye extract of pH 3.0 had the highest photocurrent density.	(Isah et al., 2015)
Red cabbage	water and ethanol	Analysis of Chameleonic Change of Red Cabbage Depending on Broad pH Range for Dye-Sensitized Solar Cells	For the red cabbage dye-sensitized electrode adsorbed at pH 3.5 corresponding to an energy conversion efficiency (η) of 0.41% which is the highest.	(Park et al., 2015)
Rosella, Bawang Sabrang, Cherry Barbados, Mulberry, Ardisia, Oxalis Triangularis and Harum Manis mango	ethanol and water	Effect of varied extracting solvent on stability and reliability of DSSCs using natural dyes as photosensitizer	The dyes extracting in water resulted in higher performance compared in ethanol solution.	(Suhaimi & Shahimin, 2014)
Monascus red	water, ethanol, and dimethylsulfoxide (DMSO)	Influence of polar solvents on photovoltaic performance of Monascusred dye-sensitized solar cell.	DSSC using natural *Monascus red* dissolved in DMSO solvent showed a higher photovoltaic performance compared to other solvents of water and ethanol.	(Lee et al., 2014)

Rosella and blue pea flowers	water and ethanol	Dye-sensitized solar cell using natural dyes extracted from rosella and blue pea flowers	The optimum extraction condition is by using ethanol as the extraction solvent, temperature of 100°C, and pH value of 1.0.	(Wongcharee et al., 2007)

2.1.6 Mangosteen Pericarp and Adenium Obesum Extraction

The efficiency of the untreated mangosteen pericarp is 1.17% based on Table 5 and the main effectual pigment that brought about such high efficiency is the rutin pigment as shown in Figure 9 which had an efficiency of 1.12% although it does not show a synergistic property with the other extracts of α-Mangostin or β-mangostin structure from mangosteen pericarp as proven in Table 2.5(H. Zhou et al., 2011).

There were several extraction methods and conditions of the mangosteen pericap researched in order to extract the desired chemical constituents for different purposes especially antioxidants chemicals in the mangosteen pericarp (Jung, Su, Keller, Mehta, & Kinghorn, 2006), (Zarena & Sankar, 2009), (H.-C. Zhou, Lin, Wei, & Tam, 2011)

Table 2.5: Mangosteen Pericarp Constituents and Its Photovoltaic Performance(H. Zhou et al., 2011).

Plant source	Structure or structural class	Photoactive area (cm^2)	J_{sc}(mA/cm^2)	V_{oc}(mV)	η(%)/FF	Remark
Mangosteen pericarp	-	0.2	2.69	686	1.17/0.63	Extract
Mangosteen pericarp	α-Mangostin/ β-mangostin	0.2	2.55	621	0.92/0.58	Fractionated extract
Mangosteen pericarp	Rutin	0.2	2.92	611	1.12/0.63	Fractionated extract

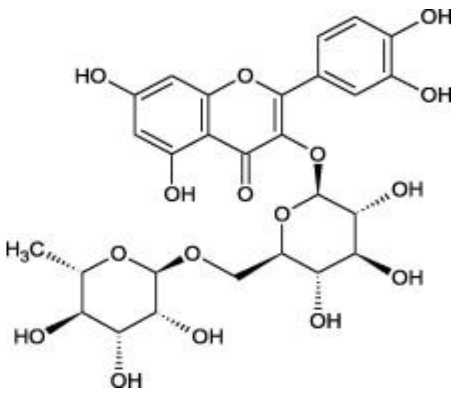

Figure 2.5 Rutin Pigment Structure of Mangosteen (Hug, Bader, Mair, & Glatzel, 2014)

Although there is no significant research on the specific pigments in adenium obesum flower, we can justify that the adenium obesum's pink colour nature is categorized under flavanoid pigment group based on the literature by Obon and Rivera in 2006. Hence, the extracted dye of adenium obesum consists mainly of flavanoid pigments as shown in Figure 2.6.

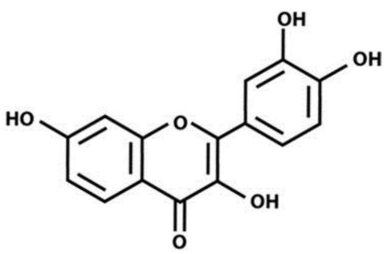

Figure 2. 6 Flavanoid Basic Chemical Structure (Shalini et al., 2015)

CHAPTER 3

METHODOLOGY

3.1 Project Flow Chart and Activities

The flow chart below shows the steps taken in order to achieve the goal of this research.

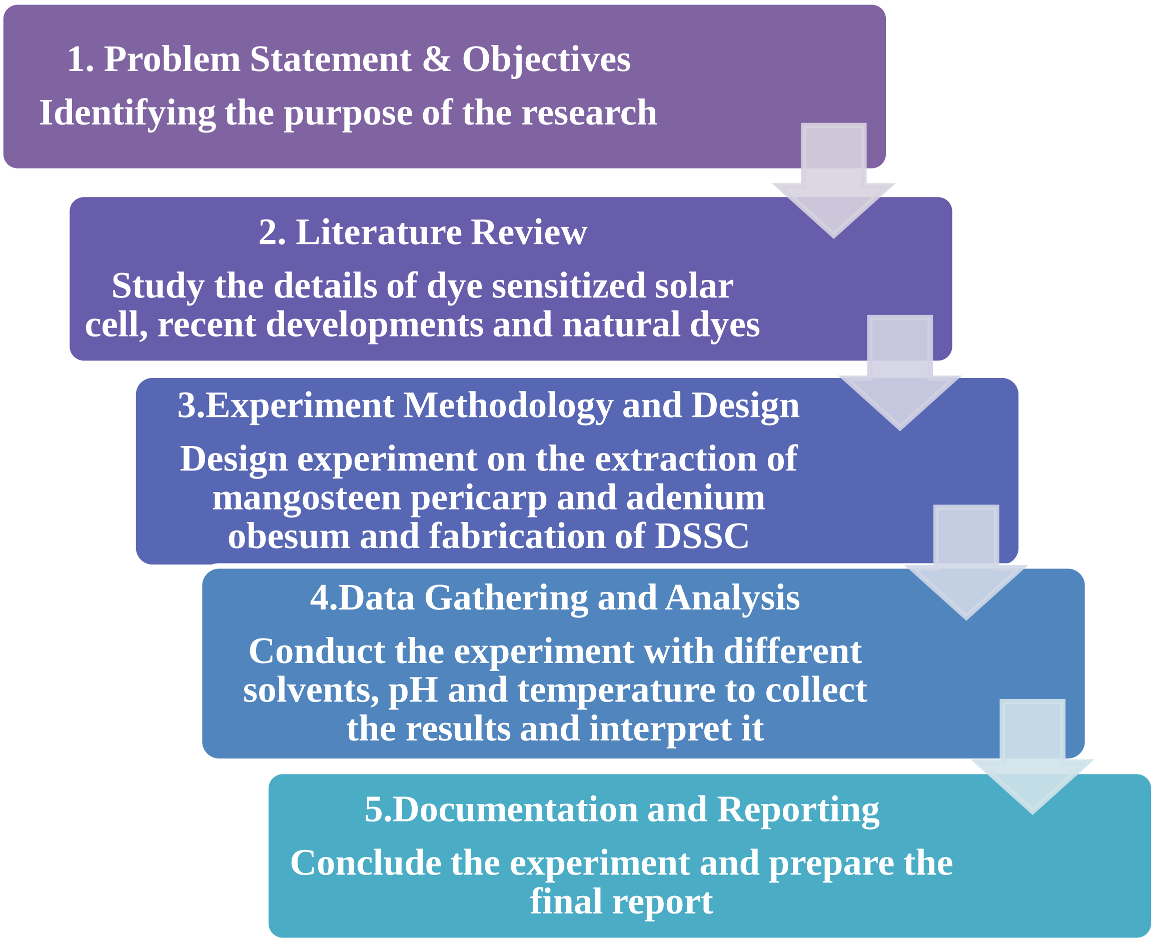

Figure 3. 1 Flow Chart

3.2 Gantt Chart and Key Milestone

Table 3.1: FYP1 Gantt Chart and Key Milestones

No	Detail/Week	1	2	3	4	5	6	7	8	9	10	11	12	13	14
1	Selection of Project Topic														
2	Preliminary Research Work														
3	Submission of Extended Proposal Defense														
4	Proposal Defense														
5	Project Work Continuation														
6	Extraction of Mangosteen Pericarp with Ethanol and Water														
7	Submission of Interim Draft Report														
8	Submission of Interim Final Report														

Process

Milestone

Table 3.2: FYP 2 Gantt Chart and Key Milestone

No	Detail/Week	1	2	3	4	5	6	7	8	9	10	11	12	13	14
1	Extraction of Mangosteen Pericarp														
2	Extraction of Adenium Obesum														
3	UV-Visible Testing of Mangosteen Pericarp with solvents														
4	UV-Visible Testing of Adenium Obesum Flower with 20°C,40°C and 60°C														
5	UV-Visible Testing for Comparisan of Mangosteen Pericarp, Adenium Obesum, and Mixture of mangosteen pericarp and adenium obesum flower														
6	Submission of Progress Report														
7	Preparation of Dye for Solar Cell Fabrication														
7	Fabrication and Assembly of Solar Cell														
8	Analysis Measurement of DSSC / Pre-SEDEX														
9	Submission of Draft Report														
10	Submission of Dissertation														
11	Submission of Technical Paper														
12	Viva Project Presentation														
13	Submission of Hard Bound Project Dissertation														

Process
Milestone
Current Progress

3.3 Experiment

3.3.1 Preparation of Natural Dye Sensitizers

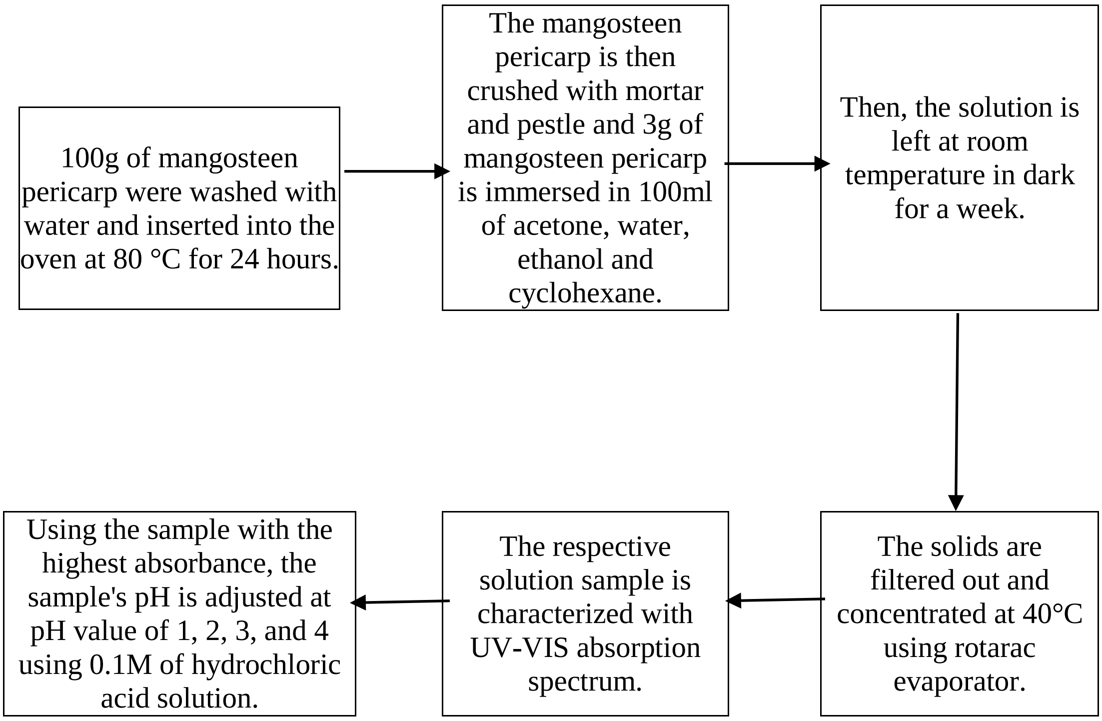

Figure 3.2 Mangosteen Pericarp Preparation and Characterization

The mangosteen pericarp was extracted with several solvents such as ethanol, acetone, chloroform, and water. First, 100g of mangosteen pericarp were washed with water and vacuum dried at 80°C. Then, the pericarp is crushed into fine powder using mortar and pestle and immersed in the respective solvents, at room temperature in the darks for a week. Then, the solids were filtered out and the filtrates were concentrated with rotary evaporator at 40°C respectively. The sample is added with hydrochloric acid to adjust the pH level as shown in Figure 3.2.

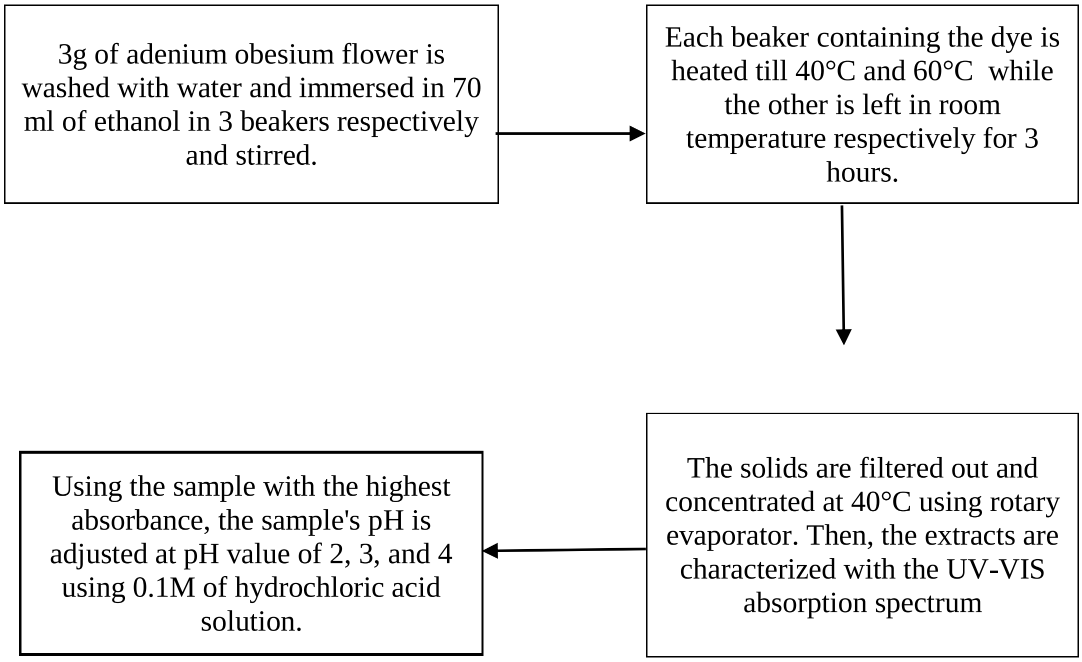

Figure 3.3 Adenium Obesum Preparation and Characterization

Then 3g of adenium obesum flower were immersed in bath water at 80°C, 90°C, and 100°C for 3 hours. Then, the solids are filtered out and each filtrate sample is added with hydrochloric acid to adjust the pH level according to Figure 3.3.

The mixture of the mangosteen pericarp and adenium obesum extract is prepared by mixing 50% of adenium obesum flower and 50% of mangosteen pericarp. Then, the mixture's pH is adjusted to several pH values less than pH 3.

3.3.2 Fabrication of DSSC

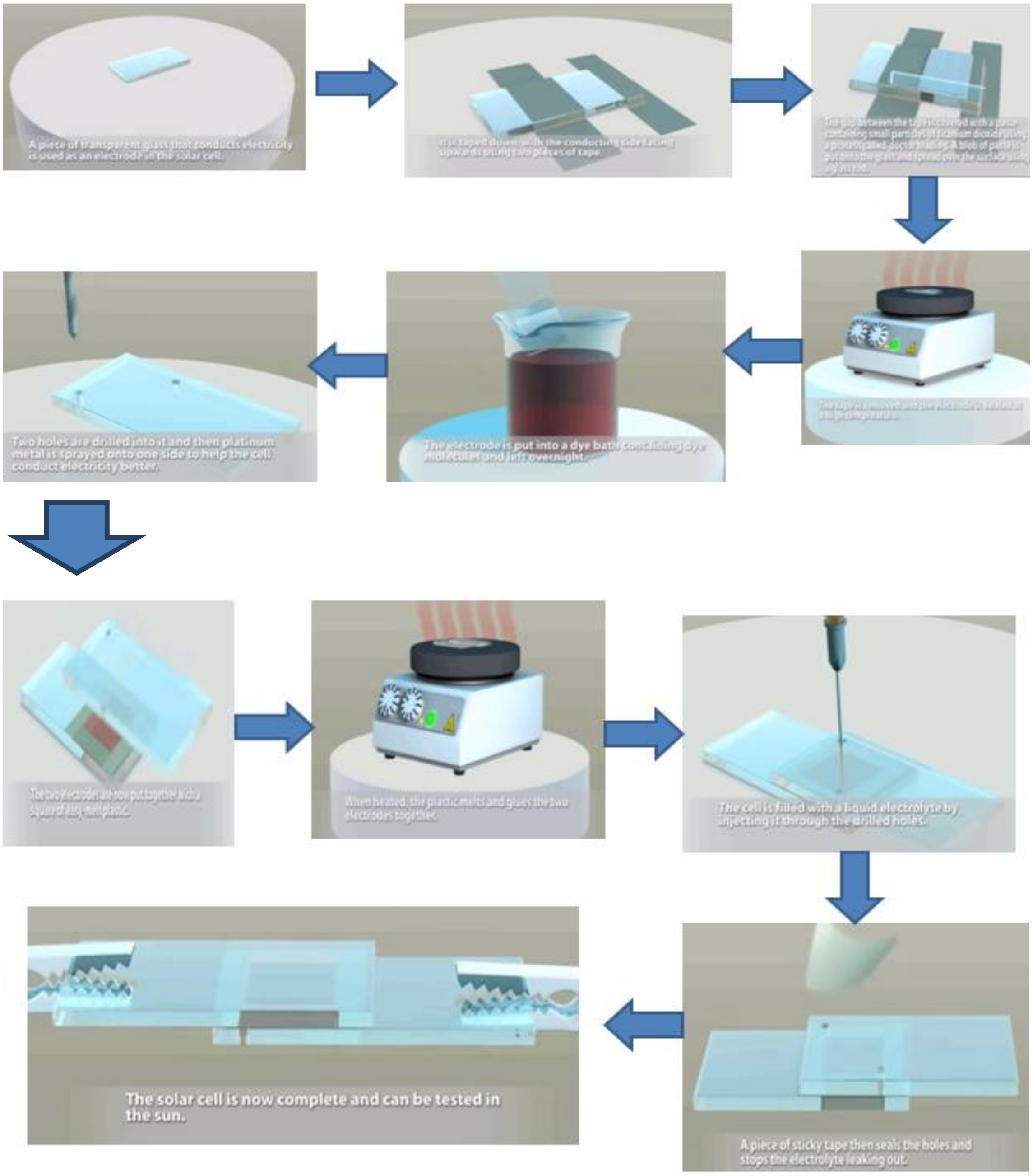

Figure 3.4 DSSC Fabrication

Conductive coated glass was obtained from the DSSC lab. A white TiO_2 paste is prepared with commercial Dyesol titanium oxide paste nanoparticles in the lab.

Two pieces of conductive glass are selected and labeled with the laser machine and the counter electrode was cut with glass cutter and drilled with the sand blasting machine. Then, both of the glasses undergoes the 'washing' process for two hours to remove the glass residues. After ‘washing’, a thin layer of titanium dioxide prepared using Dyesol 18NR-AO Active Opaque Titania Paste with thickness of 7 μm was applied twice on the photoelectrode using the screen printing method. Later, the glasses is heated at 450 °C for 1 hour in the furnace belt. Then, the conductive glass was cut into respective 1 cm^2 as per labelled. After the annealing process,the conductive glass was left to cool down to room temperature and the coated glass is immersed into the respective natural dye solution and left for 24 hours until the TiO_2 film changes colour. The excess non-adsorbed dye, were rinsed using respective solvent of the dye. Another conductive plated glass is prepared by having a layer of platinum deposited by screen-printing method and dried at 100 °C for 30 min in the drying oven belt. The thickness of Pt thin film is approximately 0.1 cm. This layer will be known as counter electrode in the solar cell configuration.

Binder clips are used to hold the conductive electrodes together. Then, the commercial electrolyte Dyesol solution which is prepared in the lab was injected and used as sandwich material between the dye coated TiO_2 layer and counter electrode material. Small amount of sealant of aluminium tape is applied around TiO_2 material area to avoid any leak of electrolyte solution. The Pt thin film was coated on ITO coated glass of the other electrode. The procedure of fabrication is as shown in Figure 3.4.

3.3.3 Characterization and Measurements

The absorption spectra of dye solutions were recorded using a UV–VIS spectrophotometer (Pelkin-Elmer, model UV-3101).

The current density-voltage (J-V) measurements were carried out under one sun illumination (AM1.5G) using a solar simulator with an intensity of 100 mWcm-2 . A Keithley 2040 was used as the source measurement unit. The photoelectric conversion efficiency (η) was calculated using the following equation: where η is the global efficiency, and V_{OC}, I_{SC} and FF are the open-circuit voltage, short circuit current density and fill factor, respectively. P_{in} is the maximum input of light energy.

$$\eta(\%) = \frac{V_{OC} \times I_{SC} \times FF}{P_{in}} \times 100 \quad (7)$$

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Effect of Natural Dye Sources On DSSC's Efficiency

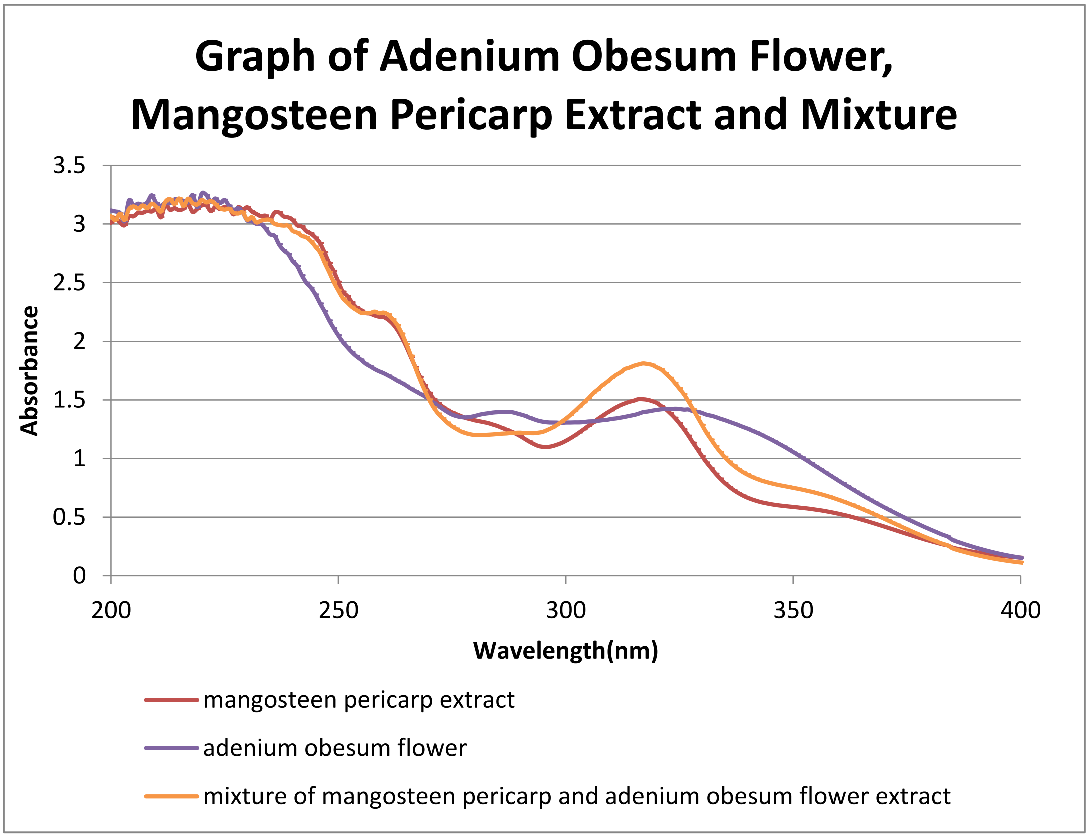

Figure 4.1 Graph of Adenium Obesum, Mangosteen Pericarp and Mixture

Figure 4.1 shows the UV-VIS absorption spectra of mangosteen pericarp extract, adenium obesum flower extract and mixture of mangosteen pericarp – adenium obesum flower. It was found that the absorption peak of the mangosteen pericarp extract is about 212 nm and 320 nm and the absorption peak of adenium obesum extract is about 220 nm and 330 nm. The absorption peak of the mixture extract is about 215 nm and 320 nm.

From the Figure 4.1 above, the extracts of mangosteen pericarp, adenium obesum and mixture shows a significant peak difference in the region of wavelength between 280- 400 nm. This might be due to the different colour, pigment group of the extracts as well as different structural group of the mangosteen pericarp and adenium obesum flower. The mangosteen pericarp extract contains the rutin group whereas the adenium obesum flower contains flavonoid structure. From the absorption spectra the mixture does show a synergistic effect. The mixture of the mangosteen pericarp and adenium obesum shows the highest absorption peak compared to the pure extracts in the region between the 280nm – 400nm region. The mixture extract absorbs more sunlight compared to the pure extracts of mangosteen pericarp and adenium obesum in the ultraviolet region. The mangosteen pericarp extract also showed a dominant influence on the absorption pattern of the mixture although the mixture has a higher absorption intensity than the pure mangosteen extract compared to the adenium obesum flower.

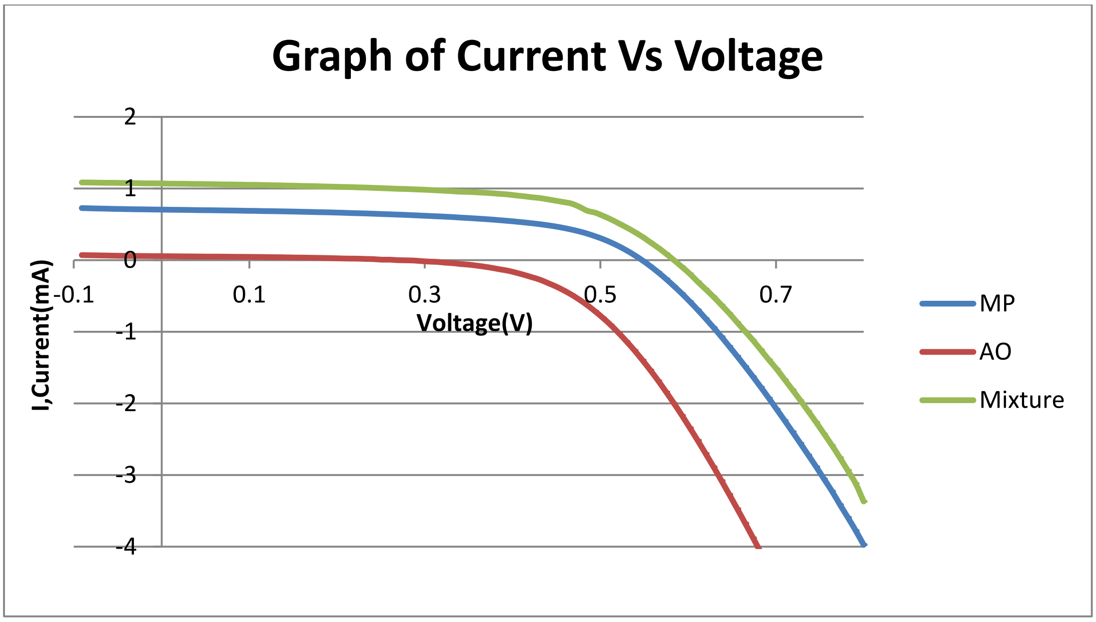

Figure 4.2 I-V Curve for Mangosteen Pericarp, Adenium Obesum and Mixture of Mangosteen Pericarp-Adenium Obesum

Table 4.1 Photoelectrochemical Parameters of Mangosteen Pericarp, Adenium Obesum, and Mixture of Mangosteen Pericarp-Adenium Obesum

Source	V_{oc} (V)	J_{sc} (mA/cm^2)	Fill Factor	Efficiency, η (%)	Efficiency comparison to literature, η (%)
Mangosteen Pericarp	0.54	0.71	0.57	0.22	0.38
Adenium Obesum	0.26	0.06	0.40	0.01	0.02
Mixture of Mangosteen Pericap -Adenium Obesum	0.58	1.07	0.60	0.37	0.64
N-719 Complex Dye (Reference)	0.63	8.27	0.31	1.78	3.09
N-719 Complex Dye (Literature) (Nursama & Muliani, 2012)	0.64	7.05	0.48	3.09	1.00

Based on Figure 4.2 and Table 4.1, the pure mangosteeen pericarp extract has a significantly higher power conversion efficiency of 0.22 % than the pure adenium obesum extracts. The structure for the rutin has more functional groups compared to the flavonoid structure to be bonded with the titanium dioxide. The mixture of mangosteen pericarp and adenium obesum showed highest power conversion efficiency of 0.37 % as compared to its pure extracts. Based on the values of open circuit voltage,V_{oc} and fill factor of the mixture and pure mangosteen extract, the mixture's photoelectrochemical performance are highly influenced by the mangosteen pericarp extract compared to the adenium obesum extract due to having a minor differences in the V_{oc} and Fill Factor value. This is in accordance with the UV-VIS absorption spectrum in Figure 4.1. The intensity of light absorbance of the dye clearly affects the photoelectrochemical performance of the DSSC.

4.2 Effect of pH Value of Extract Solution on the DSSC's Efficiency

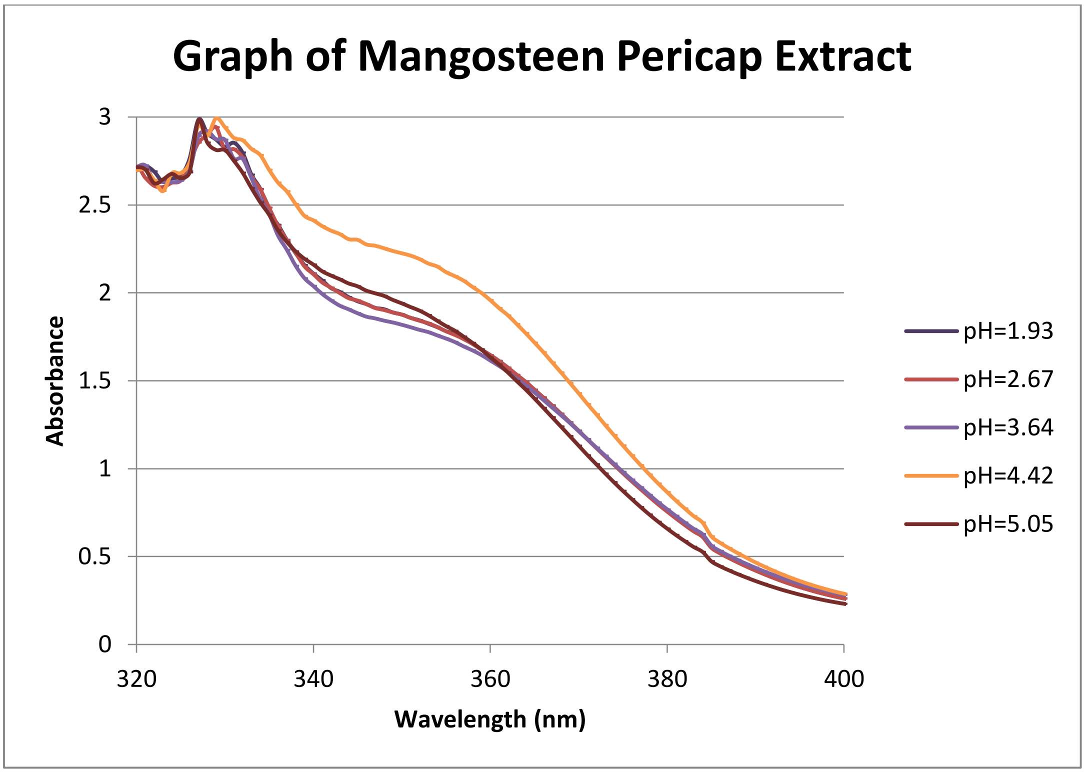

Figure 4. 3 Graph of Mangosteen Pericarp Extract

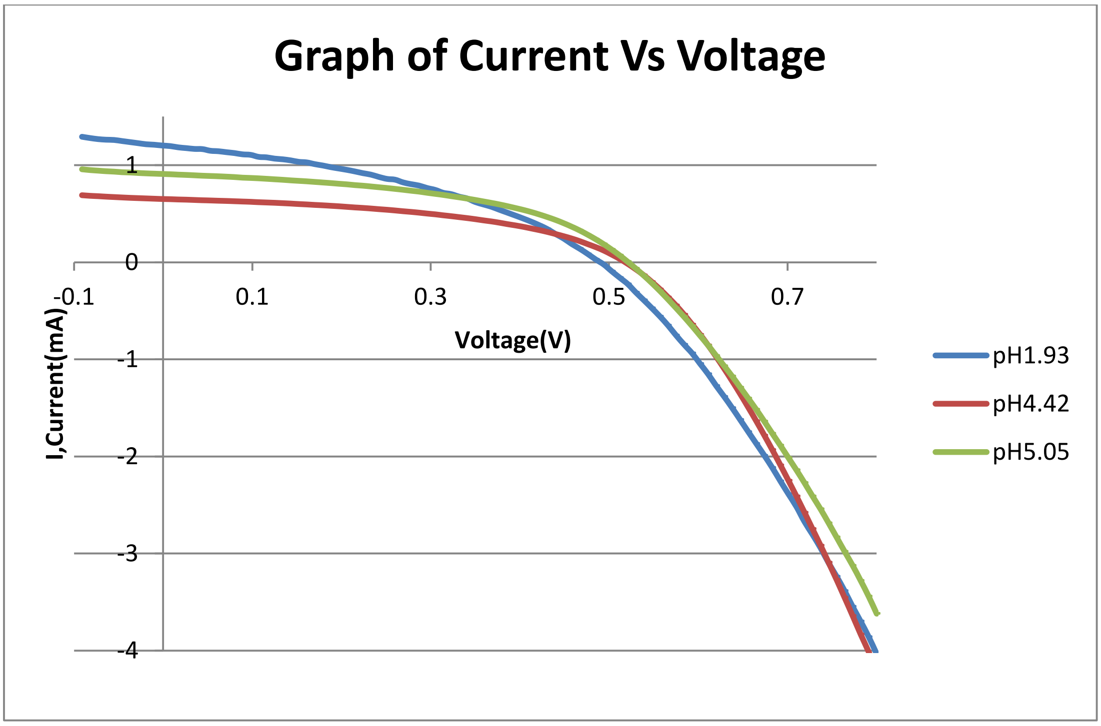

Figure 4.4 I-V Curve of Mangosteen Pericarp with pH 1.93, pH 4.42 and pH 5.05

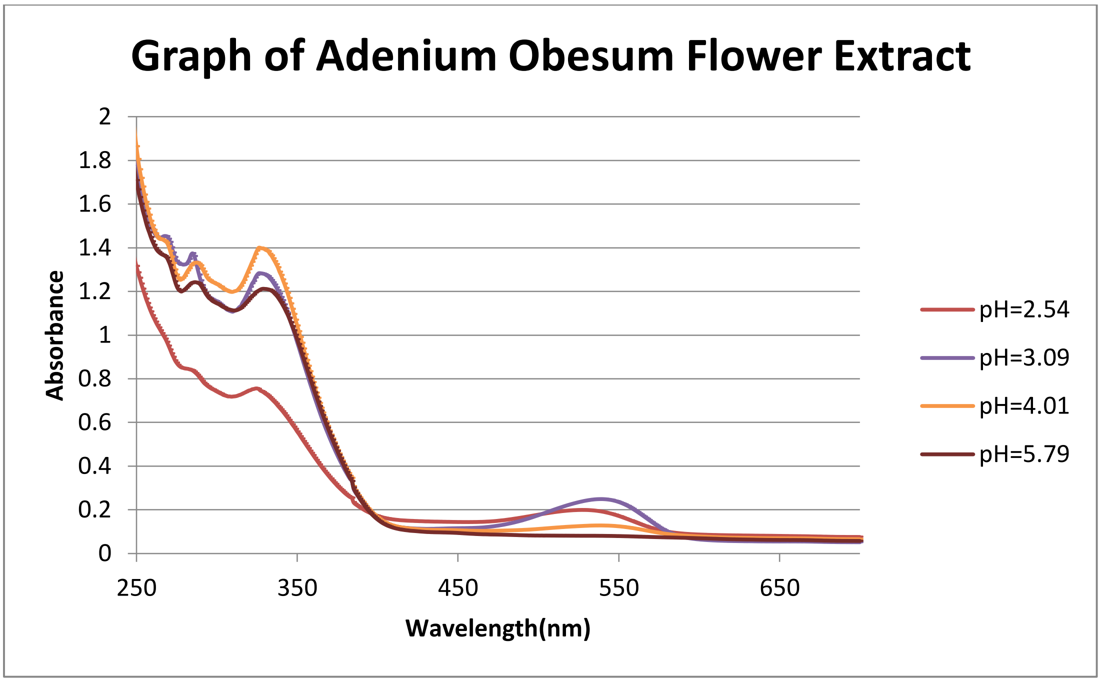

Figure 4.5 Graph of Adenium Obesum Extract

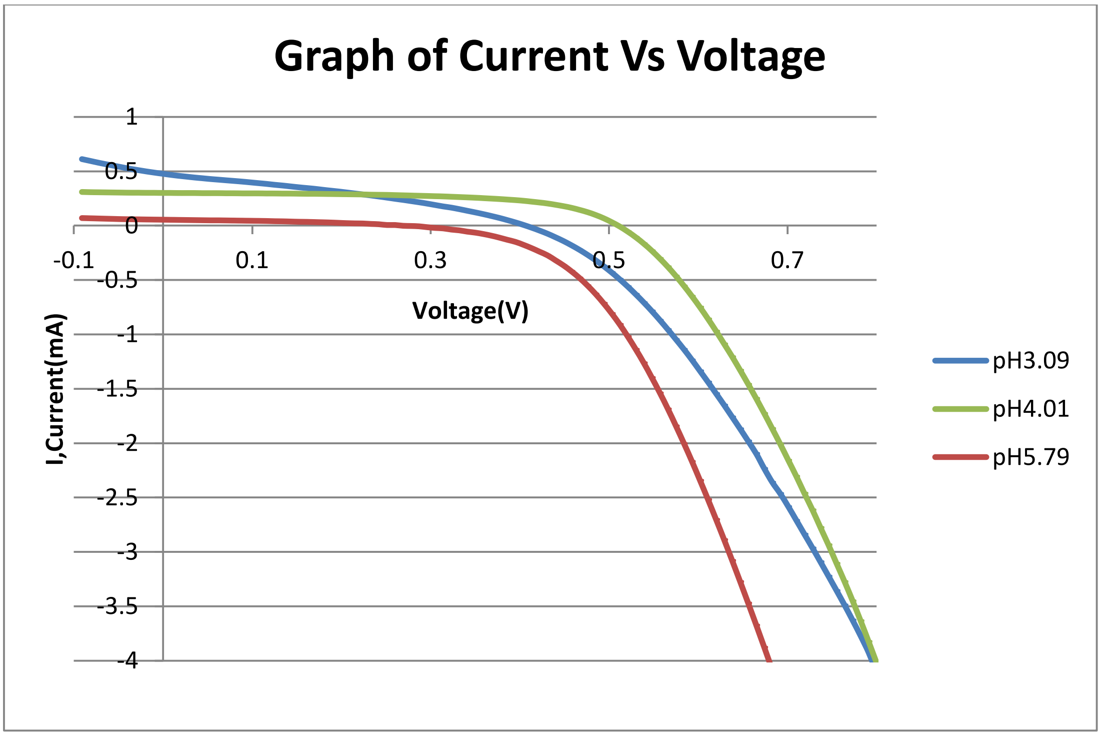

Figure 4.6 I-V Curve of Adenium Obesum Extract with pH 3.09, pH 4.01 and pH 5.79

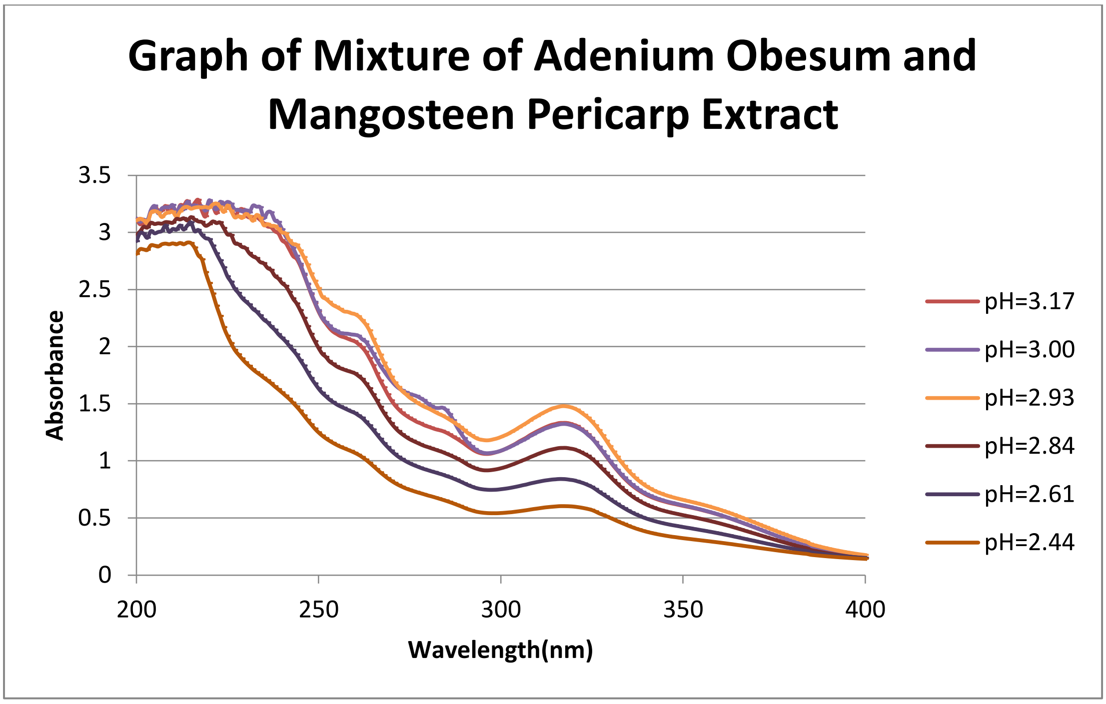

Figure 4.0.7 Graph of Mixture of Mangosteen Pericarp – Adenium Obesum Extract

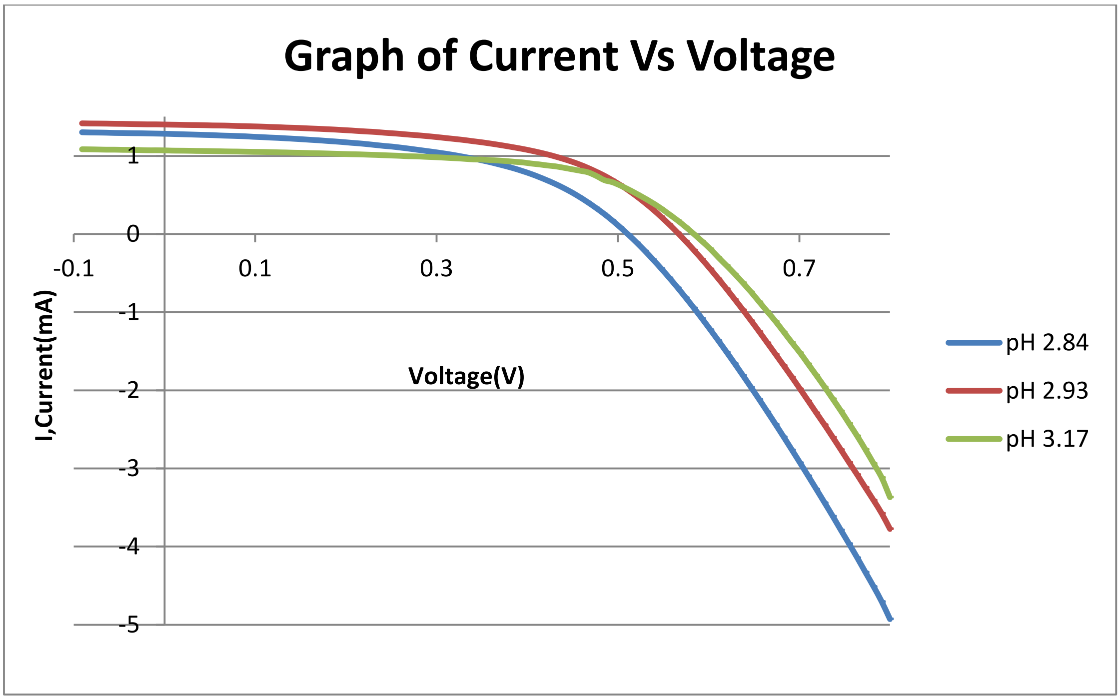

Figure 4.8 I-V Curve of Mixture of Mangosteen Pericarp-Adenium Obesum Extract with pH 2.84, pH 2.93 and pH 3.17

Table 4.2: Photoelectrochemical Parameters of Mangosteen Pericarp, Adenium Obseum and Mixture of Mangosteen Pericarp-Adenium Obesum

Mangosteen Pericarp				
pH value	V_{oc} (V)	J_{sc} (mA/cm^2)	Fill Factor	Efficiency, η (%)
pH 1.93	0.47	1.20	0.41	0.227
pH 4.42	0.51	0.65	0.47	0.156
pH 5.05	0.52	0.91	0.48	0.225
Adenium Obesum				
pH value	V_{oc} (V)	J_{sc} (mA/cm^2)	Fill Factor	Efficiency, η (%)
pH 3.09	0.40	0.48	0.35	0.065
pH 4.01	0.50	0.30	0.62	0.093
pH 5.79	0.27	0.10	0.40	0.006
Mixture of Mangosteen Pericarp and Adenium Obesum				
pH value	V_{oc} (V)	J_{sc} (mA/cm^2)	Fill Factor	Efficiency, η (%)
pH 2.84	0.51	1.28	0.50	0.330
pH 2.93	0.57	1.40	0.54	0.431
pH 3.17	0.58	1.07	0.60	0.373

The effect of pH was also investigated in this study for the mangosteen pericarp extracts, adenium obesum extracts and mixture of mangosteen pericarp – adenium obesum extracts. The original pH of the mangosteen pericarp extract was found to be 5.05. The graph in Figure 4.3 shows a significant difference peak in between wavelength of 340 nm to 360 nm. The mangosteen pericarp extract with pH 4.42 has the highest absorption peak compared to other extract solutions in between the wavelength 340 nm to 360 nm. Since the absorption spectrum for the extract with pH value of 3.64 is the lowest and the extract with pH value of 1.93 and 2.67 had a similar absorption spectrum, the extract solution with pH value of 2.67 and 3.64 was discarded for the DSSC's power conversion efficiency measurement. From the I-V curve in Figure 4.4 and Table 4.2, the mangosteen pericarp with the extract of 1.93 has the highest power conversion efficiency of 0.227 %. Compared to the mangosteen pericarp with the original pH of 5.05, the power conversion efficiency is slightly less due to having more resistance indicated in the Figure 4.4. The mangosteen pericarp extract with pH 4.42 has the lowest power conversion efficiency due to a high resistance of the circuit. The pattern is not consistent with the absorption intensity of the UV-Vis absorption spectrum graph in Figure 4.3. The DSSC performance for the mangosteen pericarp extract are independent of the pH due to the rutin structure having a high resistance towards a major pH- induced degradation (Friedman & Jürgens, 2000).

Hence, the photoelectrochemical performance for the mangosteen pericarp with pH 1.93, pH 4.42 and pH 5.05 is highly dependent on the resistance of circuit.

For the adenium obesum extract, the original pH is found to be 5.79. The graph in Figure 4.5 shows a different absorption pattern between the wavelength from 300 nm to 400 nm and from 450 nm to 600 nm. From the observation, with the addition of pH, the colour of the extract deepens from light pink to dark pink when the pH value decreases from 5.79 to 3.09. However, the colour fades to light pink when the pH value changes to 2.54. From the graph in **Figure 4.5**, the adenium obesum extract shows the highest absorption peak at the pH value of 4.01 followed by pH value of 3.09, 5.79, and 2.54 between the wavelengths of 300 nm to 400 nm. However, it shows a different pattern with the adenium obesum extract of pH value of 3.09 having the highest absorption peak followed by extract with pH value of 2.54, 4.01 and 5.79 between the wavelengths of 450 nm to 600 nm. This might be due to the effect of deepening of the pink colour as the pH decreases till 3.09. Besides that, the extract with the pH value of 2.54 was neglected in the DSSC's efficiency measurement due to its lowest absorption spectrum. From Table 4.2 and Figure 4.6, the photo conversion efficiency for the adenium obesum extract with pH 4.01 is the highest compared to the extract with pH 3.09 and pH 5.79. It correlates with the UV-VIS absorption spectrum in Figure 4.5. The intensity of the light absorbance of the adenium obesum from 300 nm to 400 nm dominates the power conversion efficiency of the DSSC instead of light absorbance wavelength between 450nm to 600nm. By reducing the pH from 5.79 to 4.01 value, the efficiency is increase by a multiplication of 15.5. The anthocyanin in the flavonoid structure hydrolyses to the neutral quinonoidal bases as seen in Figure 4.9 which is stable and has a stronger bonding with the titanium dioxide at the pH 4.01. (Jackman & Smith, 1996). The dye pH of adenium obesum has a major impact to its DSSC photo conversion efficiency.

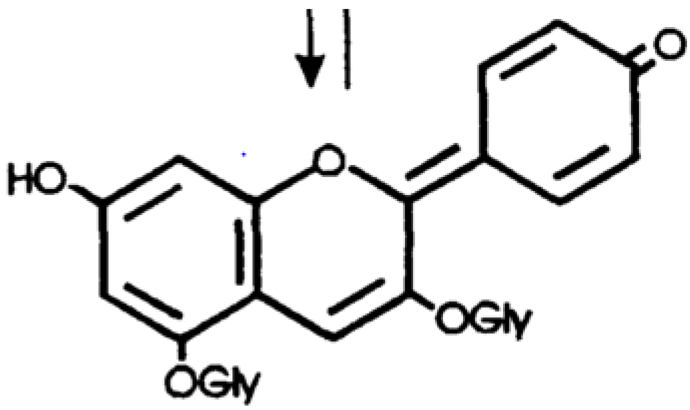

Figure 4.9 Neutral Quinonoidal Bases Structure

For the mixture of mangosteen pericarp and adenium obesum extract, the original extract pH was found to be at 3.17. From the graph in the Figure 4.7, it shows a similar absorption pattern but different absorbance peaks. The mixture extract with the pH value of 2.93 has the highest absorbance peak followed by the extract with pH value of 3.00, 3.17, 2.84, and 2.61. The mixture extract with pH value of 2.44, 2.61 and 3.00 was neglected in the DSSC's efficiency measurement due to low absorption spectrum and having similar absorption spectrum of extract for pH value of 3.00 and 3.17 in the UV-VIS characterization. Based on the Table 4.2, the significant dominant photoelectrochemical parameter which affects the photo conversion efficiency is the short circuit current parameter for the mixture of the mangosteen pericarp and adenium obesum extract. The mixture with pH value of 2.93 had the highest efficiency value followed by pH 3.17 and pH 2.84. The light absorption of the mixture with various pH affects the short circuit current value of the DSSC.

4.3 Effect of Extracting Solvents on DSSC's Efficiency

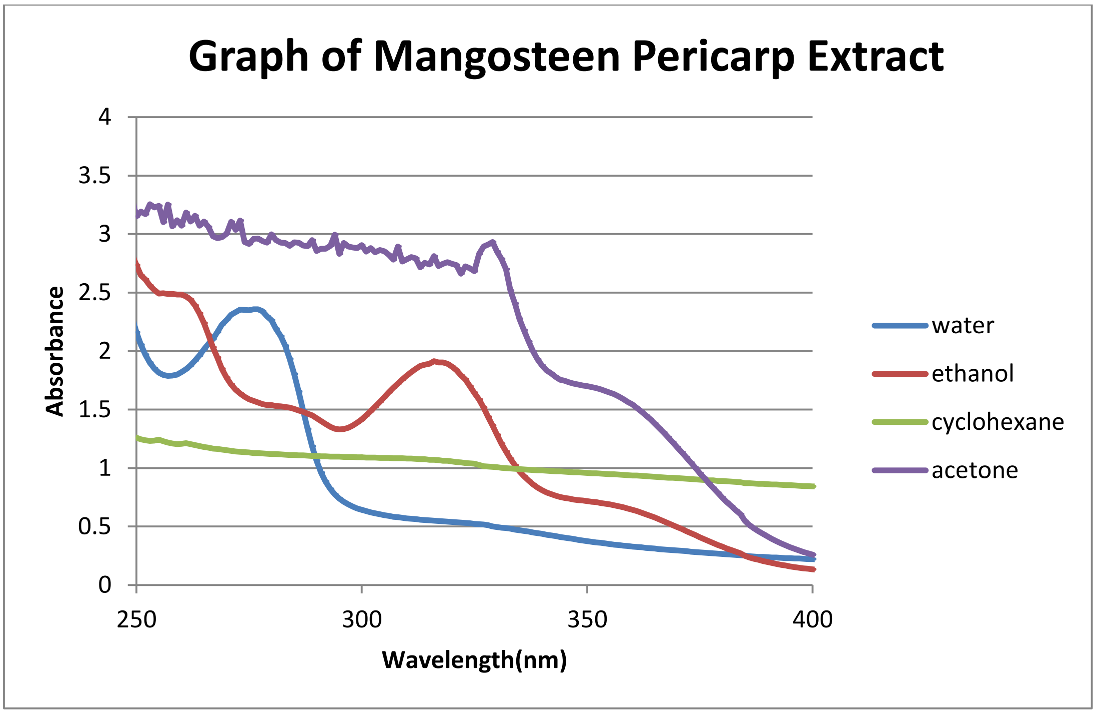

Figure 4.10 Graph of Mangosteen Pericarp Extract

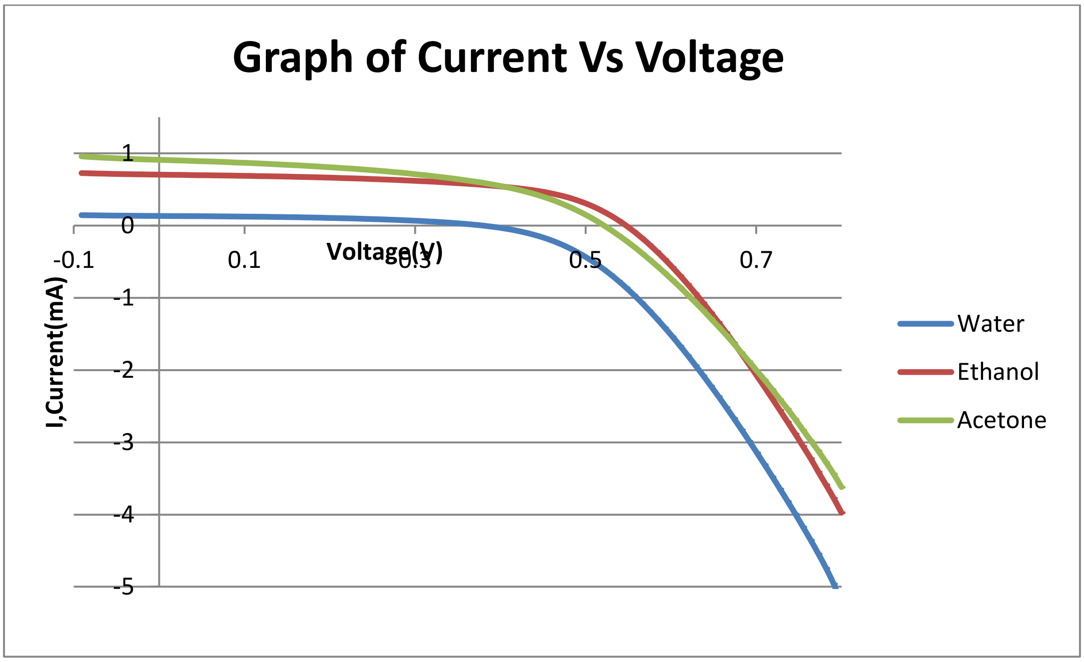

Figure 4.11 I-V Curve of Mangosteen Pericarp Extracted with Water, Ethanol and Acetone

Table 4.3: Photoelectrochemical Parameters with Mangosteen Pericarp Extracted with Water, Ethanol, and Acetone

Solvent	V_{oc} (V)	J_{sc} (mA/cm^2)	Fill Factor	Efficiency, η (%)	Efficiency comparison to literature, η (%)
Water	0.37	0.13	0.45	0.02	0.03
Ethanol	0.54	0.71	0.57	0.22	0.38
Acetone	0.52	0.91	0.48	0.23	0.40
N-719 Complex Dye(Reference)	0.63	8.27	0.31	1.78	3.09
N-719 Complex Dye(Literature) (Nursama & Muliani, 2012)	0.64	7.05	0.48	3.09	1.00

The effect of extracting solvent was investigated in the extraction of mangosteen pericarp. From the graph 4.9, the mangosteen pericarp extracted using acetone showed the highest absorption peak of compared to other extracting solvent. The mangosteen pericarp is soluble in acetone solvent compared to the other solvent. The mangosteen pericarp extracted using the water showed highest absorption peak at 280 nm while the ethanol extract show a peak at 320 nm. However, the mangosteen pericarp extracted using the cyclohexane solvent did not show any peak which indicates that cyclohexane is not suitable as an extracting solvent for mangosteen pericarp. From the Figure 4.9, the extract with cyclohexane solvent was neglected in the DSSC‘s efficiency measurement due to its bad absorption spectrum. From the Figure 4.10, the mangosteen pericarp extracted with acetone has the highest efficiency of 0.23 %. The I-V curve of mangosteen pericarp extracted with acetone shows less resistance than the I-V curve of mangosteen pericarp extracted with ethanol which helps to increase the short circuit current value as well as the efficiency. The mangosteen pericarp extract is insoluble in non polar solvent such as cyclohexane. The extract is highly soluble in the polar aprotic solvent such as acetone compared to the polar protic solvent such as water and ethanol. As seen with the ideal metal complex dye, N-719 dye shows close values of open circuit voltage, V_{oc} and fill factor with the natural dye extracted from mangosteen pericarp. Hence, the short circuit current is highly influenced by the type of dye used for the DSSC.

4.4 Effect of Extraction Temperature of DSSC's Efficiency

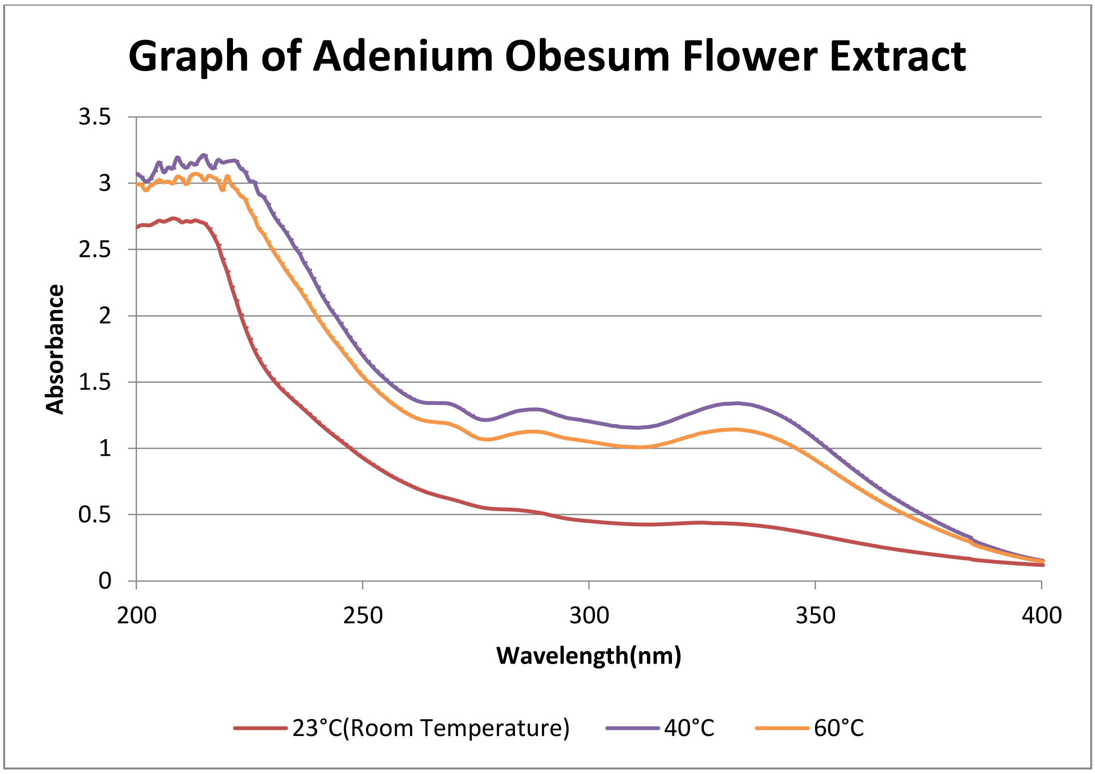

Figure 4.12 Graph of AdeniumObesum Extract

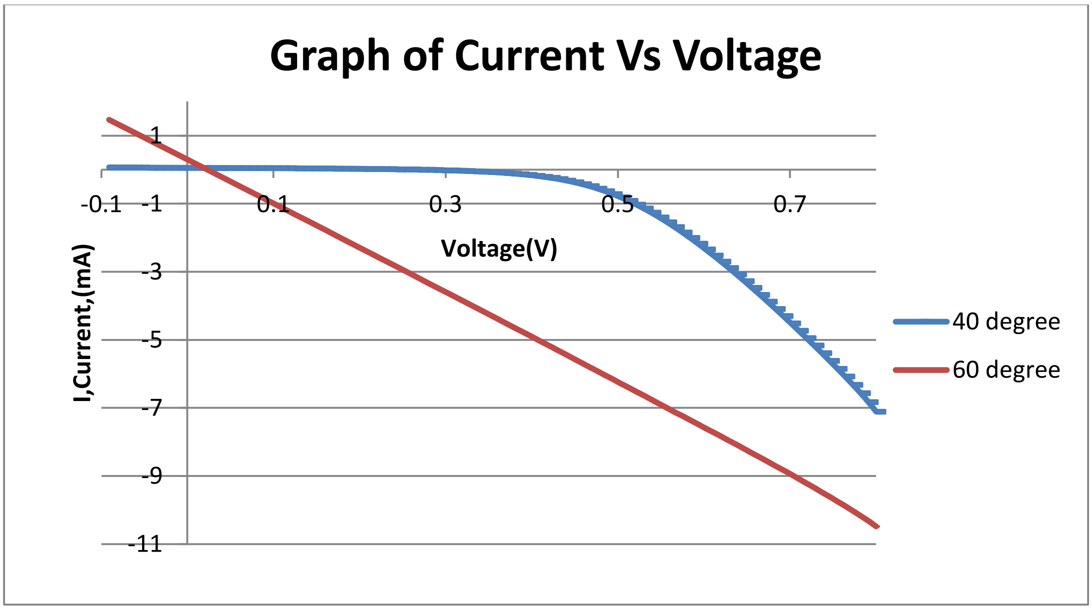

Figure 4.13 Graph of I-V Curve of Adenium Obesum Extracted at 40°C and 60°C

Table 4.4: Photoelectrochemical Parameters of Adenium Obesum Extracted at 40°C and 60°C

Temperature	V_{oc} (V)	J_{sc} (mA/cm^2)	Fill Factor	Efficiency, η (%)	Efficiency comparison to literature, η (%)
40ºC	0.26	0.05	0.40	0.006	0.010
60ºC	0.02	0.31	0.22	0.002	0.003
N-719 Complex Dye(Reference)	0.63	8.27	0.31	1.780	3.090
N-719 Complex Dye(Literature)	0.64	7.05	0.48	3.090	1.000

The extraction temperature was investigated using the adenium obesum extract. From the graph of Figure 4.11, it can be concluded that the optimum temperature for the extraction of adenium obesum flower is 40°C as it shows the highest absorbance peak compared to 60°C and 23°C. Hence, the adenium obesum extract of 23°C was discarded in the DSSC's efficiency measurement due to its low absorption spectrum in Figure 4.11. From the Figure 4.12 and table 4.4, the efficiency of the adenium obesum extracted at 40°C has a higher efficiency compared to 60°C. The absorption intensity in the UV-Vis spectrum affects the photoelectrochemical properties. The 40°C is the optimum temperature in extracting the adenium obesum. At higher temperature of 60°C, the adenium obesum structure deteriorates quickly causing the extraction quality decreases.

CHAPTER 5

CONCLUSIONS AND RECOMMENDATIONS

As shown in the research, the dye sensitized solar cell is one of the innovative technology to provide an environmental friendly alternative for energy source.

Natural dyes provide promising alternative for a lower preparation cost, environmental friendly, and non-toxic as well as abundantly available. Although there were several factors that led to the low efficiency of natural dye, the tailoring of extraction condition in the extraction of pigments might be able to improve the performance of dye sensitized solar cell. Hence, the influence and behavior of various extracting condition of natural dyes had to be thoroughly investigated for the mangosteen pericarp dyes and adenium obesum flower dye to achieve the optimized efficiency level. From this study, the mangosteen pericarp extracted with acetone solvent shows the highest light absorption intensity and PCE value compared to the other solvent. The optimum pH condition for the mangosteen pericarp extract is pH 1.93. For the adenium obesum extract, the appropriate temperature is at 40ºC and extracted at pH 4.01. The mixture shows a higher light absorption intensity and PCE values compared to its pure extracts. The optimum pH for the mixture extract is at pH 2.93. We have also shown that the mixture of two natural dyes provide synergistic effect on the light absorption spectrum and power conversion efficiency. The best DSSCs is obtained with the mixture of mangosteen pericarp and adenium obesum flower dye in ethanol, at a pH of 2.93 with a power conversion efficiency of 0.431 %. It is actually recommended to conduct a further research for the ways for the improvement of the dye of the mixture of adenium obesum and mangosteen pericarp's aggregation and stability as well as the DSSC's power conversion efficiency.

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APPENDICES

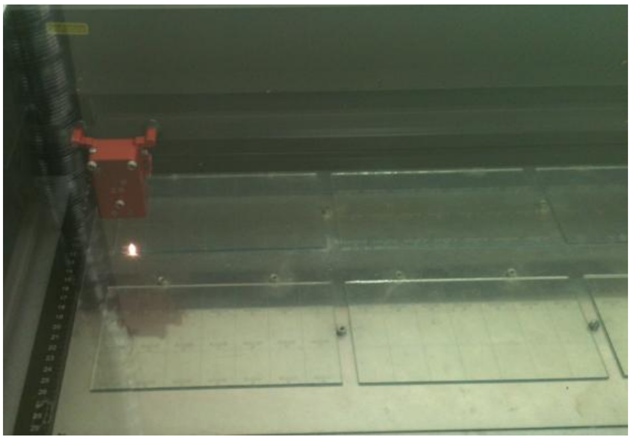
a. Mangosteen Pericarp Extract with Water, Acetone, Ethanol and Cyclohexane (from left to right) Solvent

b. Adenium Obesum Extract with Ethanol Solvent at 23°C, 40°C and 60°C (from left to right)

c. Diluted Adenium Obesum Extract with pH value of 4.01,3.09 and 2.54 (from left to right)

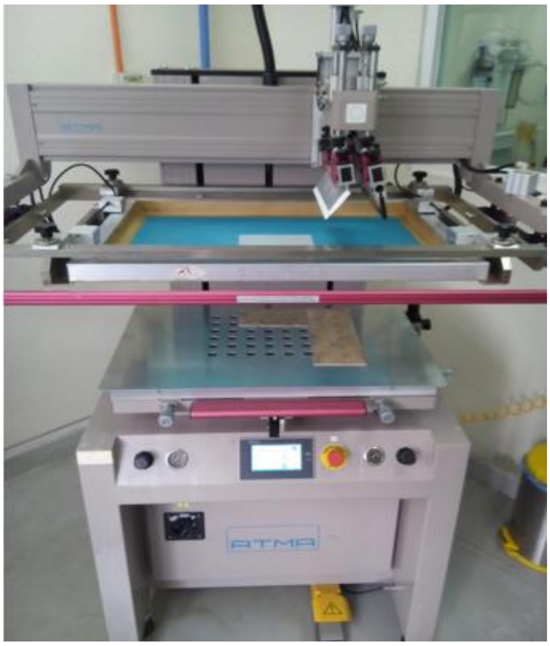
d. Mangosteen Pericarp Extract Concentrated with Rotarac Evaporator

e. Labelling of the Photoelectode and Counter Electrode with Laser Machine



f. Counter Electrode Drilled with Sand Blasting Machine

g. Titanium Dioxide Applied on the Photoelectrode with Screen Printing



h. Photoelectrode sintered in oven after screen printing of titanium oxide

i. Photoelectrode soaked with dye and titanium oxide coloured.