

**EXPERIMENTAL STUDIES AND CONSUMERS'  
WILLINGNESS TO PAY ON BIODEGRADABLE AND NON-  
BIODEGRADABLE MATERIALS IN ELECTROCHEMICAL  
INDUSTRY**

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KUALA LUMPUR**

**2018**

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NON-BIODEGRADABLE MATERIALS IN  
ELECTROCHEMICAL INDUSTRY**

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**EXPERIMENTAL STUDIES AND CONSUMERS' WILLINGNESS TO PAY  
ON BIODEGRADABLE AND NON-BIODEGRADABLE MATERIALS IN  
ELECTROCHEMICAL INDUSTRY**

**ABSTRACT**

In this research, two types of polymer electrolytes were prepared to obtain biodegradable and non-biodegradable polymer electrolytes. Poly (vinyl alcohol) (PVA) was selected as the biodegradable polymer which was incorporated with magnesium triflate  $[\text{Mg}(\text{CF}_3\text{SO}_3)_2]$  as salt and 1-butyl-3-methylimidazolium bromide (BmImBr) as the ionic liquid. Polymer electrolyte films comprising poly (ethyl methacrylate) (PEMA) as host,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  as salt and BmImBr as the ionic liquid were prepared, classified under the non-biodegradable polymer electrolyte. Differential Scanning Calorimetry (DSC) was carried out on both types of polymer electrolytes to investigate the glass transition temperature ( $T_g$ ) which was used to study the plasticizing effect of the ionic liquid. The PVA based polymer electrolyte with 60 wt.% of BmImBr gave the highest ionic conductivity at  $(1.64 \pm 0.01) \times 10^{-3} \text{ Scm}^{-1}$ . Highest ionic conductivity of  $(1.80 \pm 0.01) \times 10^{-4} \text{ Scm}^{-1}$  was obtained for PEMA based polymer electrolyte with 40 wt% ionic liquid at ambient temperature. These results showed that biodegradable based polymer electrolytes exhibited higher ionic conductivity compared to non biodegradable based polymer electrolytes at room temperature. Temperature dependent ionic conductivity studies were performed to elucidate the mechanism of ion transport and it was found that both the systems obeyed the Vogel-Tamman-Fulcher (VTF) mechanism. X-ray diffraction (XRD) investigation revealed that both the biodegradable and non-biodegradable polymer electrolytes displayed excellent complexation. The highest conducting ionic liquid based polymer electrolyte for both polymers was subsequently used to fabricate Electrical Double Layer Capacitors (EDLC). Electrochemical capacitance of the EDLC was evaluated using Linear Sweep Voltammetry (LSV),

Cyclic Voltammetry (CV) and Galvanostatic Charge Discharge (GCD). CV proved the improvement in specific capacitance of the EDLC containing ionic liquid-added polymer electrolyte. The specific capacitance of EDLCs obtained in CV was in good agreement with GCD curve. Biodegradable based EDLC showed a specific capacitance of  $45.95 \text{ Fg}^{-1}$  and non-biodegradable based EDLC displayed a capacitance value of  $7.34 \text{ Fg}^{-1}$ . Once again the better performance of the biodegradable based electrochemical device was proven through the study. In the second part of the study, a survey was conducted on consumers' willingness to pay for 'green cellphones' and how much they would be willing to pay for the device compared to the conventional phones. Findings suggest that majority of the respondents are willing to pay for the green cellphone with varying amount. From the survey carried out, we can conclude that the significant determinants that contribute to Malaysian willingness to pay for green electronics are peer pressure, health and safety, occupation, ethnicity and age.

Keywords: Poly(vinyl alcohol), Poly(ethyl methacrylate), Ionic liquid,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ , Willingness to pay

**KAJIAN EKSPERIMEN DAN KESANGGUPAN PENGGUNA MEMBAYAR  
BAGI BAHAN BIODEGRADASI DAN BUKAN-TERBIODEGRADASI DALAM  
INDUSTRI ELEKTROKIMIA**

**ABSTRAK**

Dua jenis elektrolit polimer telah disediakan untuk mendapatkan elektrolit polimer mesra alam dan tidak mesra alam. Poli (vinil alkohol) (PVA) telah dipilih sebagai polimer mesra alam yang diperbadankan dengan garam Magnesium Triflate [ $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  atau MgTf] dan 1-butyl-3-methylimidazolium bromida (BmImBr) sebagai cecair ionik. Filem elektrolit polimer yang terdiri daripada poli (etil metakrilat) (PEMA) sebagai hos, garam Magnesium Triflate [ $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  atau MgTf] dan 1-butyl-3-methylimidazolium bromida (BmImBr) sebagai cecair ionik disediakan, yang dikelaskan di bawah polimer elektrolit bukan mesra alam. Differential Scanning Calorimetry (DSC) dijalankan ke atas kedua-dua elektrolit polimer untuk menyiasat suhu peralihan kaca yang digunakan untuk mengkaji kesan pemplastikan cecair ionik. Polimer elektrolit dengan cecair ionik menunjukkan suhu peralihan kaca ( $T_g$ ) lebih rendah daripada polimer elektrolit yang bebas dari cecair ionik yang membuktikan sifat amorfus elektrolit polimer yang mengandungi cecair ionik. Polimer elektrolit PVA berasaskan dengan 60 wt.% BmImBr memberikan kekonduksian ionik yang paling tinggi di  $(1.64 \pm 0.01) \times 10^{-3} \text{ Scm}^{-1}$ . Bagi PEMA pula, kekonduksian ionik tertinggi  $(1.80 \pm 0.01) \times 10^{-4} \text{ Scm}^{-1}$  diperolehi dengan 40% berat cecair ionik pada suhu bilik. Keputusan ini menunjukkan bahawa elektrolit polimer biodegradasi menunjukkan kekonduksian ionik yang lebih tinggi berbanding dengan elektrolit polimer bukan biodegradasi pada suhu bilik. Kajian kekonduksian ionik lawan suhu dilakukan untuk menjelaskan mekanisme pengangkutan ion dan didapati bahawa kedua-dua sistem mengikut mekanisme Vogel-Tamman-Fulcher (VTF). Penyiasatan XRD menunjukkan

bahawa kedua-dua elektrolit polimer biodegradasi dan tidak terbiodegradasi menunjukkan pengkompleksan yang sangat baik. Polimer elektrolit yang menunjukkan kekonduksian tertinggi digunakan untuk merekabentuk Electric Double Layer Capacitors (EDLC). Kemuatan elektrokimia EDLC dinilai dengan menggunakan Linear Sweep Voltammetri (LSV), Cyclic voltammetri (CV) dan caj discaj Galvanostatic (GCD). Cyclic voltammetry (CV) membuktikan peningkatan dalam kemuatan khusus EDLC mengandungi ion cecair tambah polimer elektrolit. GCD tidak dapat dilakukan bagi kerana elektrolit polimer PEMA disebabkan rintangan dalaman yang tinggi. Dalam bahagian kedua, kajian telah dijalankan ke atas kesanggupan pengguna untuk membayar untuk 'telefon bimbit hijau' dan berapa banyak mereka bersedia untuk membayar bagi 'telefon bimbit hijau' berbanding yang konvensional. Keputusan kajian membuktikan bahawa kebanyakan responden sanggup membayar amaun yang berbeza untuk membeli telefon bimbit hijau. Dari kaji selidik yang dijalankan, kita dapat menyimpulkan bahawa penentu penting yang menyumbang kepada kesediaan rakyat Malaysia untuk membayar bagi elektronik hijau khususnya telefon bimbit hijau adalah tekanan rakan, kesihatan dan keselamatan, bidang kerja, etnik dan umur.

Kata kunci: Poly(vinyl alcohol), Poly(ethyl methacrylate), cecair ionic,  $Mg(CF_3SO_3)_2$ , kesanggupan membayar

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## LIST OF SYMBOLS AND ABBREVIATIONS

LITFSI	:	Lithium bis (trifluoromethanesulfonyl) imide
Mg(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	:	Magnesium trifluoromethanesulfonate
BmImBr	:	1-ethyl-3-methylimidazolium bromide
CH <sub>3</sub> COONH <sub>4</sub>	:	Ammonium acetate
PVA	:	Poly (vinyl alcohol)
PEMA	:	Poly (ethyl methacrylate)
LiPF <sub>6</sub>	:	lithium hexafluorophosphate
BmImPF <sub>6</sub>	:	1-butyl-3-methylimidazolium hexafluorophosphate
EMITf	:	1-ethyl-3-methylimidazolium trifluoromethanesulfonate
PMA	:	phosphomolybdic acid
BMITFSI	:	1-butyl-3-methylimidazolium Bis (trifluoromethanesulfonyl) imide
EMI-BF <sub>4</sub>	:	1-ethyl-3-methylimidazoliumtetrafluoroborate
VOC	:	Volatile organic compounds
SPE	:	Solid polymer Electrolyte
ECA	:	Electrochemical applications
IoT	:	Internet of Things
WEEE	:	Waste electrical and electronic equipment
EDLC	:	Electric double layer capacitors

## CHAPTER 1: INTRODUCTION

This chapter gives an overview of the topics that will be discussed in the thesis and the motivations for this study.

This study comprises two main parts, the experimental and the survey. In the first part, polymer electrolytes, both biodegradable and non-biodegradable, are prepared and characterized. Following that, both the polymer electrolytes are used to fabricate an electrochemical device, in this case the electric double layer capacitor (EDLC). The device will be characterized to determine the strengths and the weaknesses of the biodegradable and non-biodegradable polymer electrolytes. The second part of the study involves a survey among working consumers on willingness to pay for green electronics.

The research question would describe the reason to conduct the study followed by the objectives that need to be achieved. Finally, the novelty of the work would be described and in conclusion, the scope of the thesis would be explained.

### **1.1 Malaysia and Going Green**

As the world moves towards green energy and sustainability, Malaysia as a developing country refuses to be left behind. The Malaysian government has committed itself to move towards sustainability and green lifestyle, however various industries in Malaysia, including the manufacturing and electrical and electronic industry are contributing towards the carbon dioxide emission which causes harm to the environment. In 2013, total amount of CO<sub>2</sub> emissions from manufacturing industries was 53 million metric tonnes or 20 percent of total CO<sub>2</sub> emission in Malaysia. Having

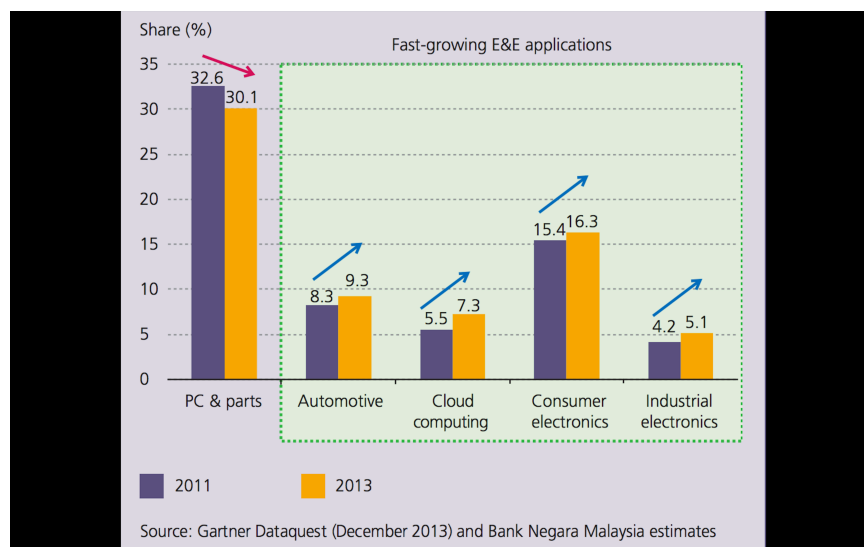
said that, the Malaysian government has pledged to reduce carbon emission up to 40 percent by the year 2020, compared to the carbon emission levels recorded in 2005. This was pledged by the Prime Minister at the Conference of the Parties (COP 15) in Copenhagen in 2009. Following that, the government reported that they had already achieved 33% in 2015. This means Malaysia needs to implement towards greener economy ensuring sustainability and safety without compromising the development of its industries. Due to these reasons, the scientific community in Malaysia has embarked on the 'green' concept to assist the policy makers in moving towards these goals.

One question which can be asked is “Where was green Technology all this while?” and “Why the sudden hype on Green Technology”? Green Technology is actually an outcome of global warming. Due to the natural disasters and climate change issues caused by rising temperatures, flash flooding, sudden earth quakes and mass deforestation are motivations for the need of Green Technology. Malaysia is also a victim of such disasters such as floods and haze which we believe is a result of global warming as well (Yaakob, 2011).

Green Technology can be defined as the use of technology of any kind associated with green chemistry, environmental friendliness, environmental science and electronics to ensure the conservation of the environment and sustainability of its resources. The area of sustainable economic development has been deemed essential in Malaysia's long-term development plans and green technology has been incorporated into the national economic growth as one of the key drivers.

## 1.2 Electrical and Electronics Industry in Malaysia

Figure 1.1 shows the share of Malaysia's electric and electronics component sales. The electric and electronics industry in Malaysia maintains itself at the pinnacle as a key driver of the Malaysian industrial development and significantly contributes to GDP growth, earnings through export, investment and employment. For example, in 2015, the value of electric and electronics export increased 8.5% to RM 277.92 billion from RM 256.14 billion in 2014 (Qader, 2011).



**Figure 1.1: Share of Malaysia's Electrical and Electronic Component Sales**

*(Source: Gartner Dataquest (December 2013) and Bank Negara Malaysia estimates)*

However, the industry has faced several challenges and had to make a paradigm shift in global preferences to reach at its level now. As seen in Figure 1.1, from an initial market of traditional personal computers (PCs) and parts manufacturers, as the world moved more fervently towards more modern smart devices and wireless technology, Malaysia's industry had to take the backseat while still trying to remain competitive in the global market. The salvation came in the form of fast-growing and

higher value-added segments. To name a few would be the demand for automotive sensors and the demand for data storage through internet related activities. Malaysia is further diversifying its market in the effort to capitalise the market demand for new applications of wireless communications and wearable devices. Known colloquially as the “Internet of Things” (IoT) – ranging from wearables, common electrical appliances as well as bio-medical devices, it presents exciting opportunities for the electrical and electronics industry in Malaysia (Malaysia, 2016). Having said that, as one of the largest industries in Malaysia, the electrical and electronics industry is also accountable as one of the largest contributors to water pollution, air pollution and solid waste (Qader, 2011). These factors further intensity the need for green practices.

Globalization has put forward new and fresh demands in the field of electric and electronics and Malaysia, to remain in the market front, has to ensure its compliance. The changing landscape in the business world and consumer demands is making industries re-invent themselves by implementing new strategies in line with going green.

### **1.3 Biodegradable and Non-biodegradable Materials in Electrochemical Devices for Environment Sustainability**

It is important to promote green technology and green practices in industries as this would lead to safer products and ensure environmental sustainability. In addition, it is crucial for businesses to realize the importance of sustainability to remain competitive in the global market.

Be it in the manufacturing or electronics industry, innovation is necessary to find ways of manufacturing green or environmentally friendly products. One such industry is

the furniture industry in Malaysia, which has taken seriously the mandate to go green as outlined by the government. The Malaysian Furniture Promotion Council has outlined several criteria to define green Malaysian furniture and one such criteria is that the wood and fabric used must be from sustainable resources and that any usage of chemicals should wholly conform to health and safety regulations and requirements. Producing green products whether it is furniture or electronics would root from the base materials used. Using sustainable or biodegradable materials would ensure that the product is also environmental friendly. A material that is non biodegradable is usually made from non-sustainable constituents. The negative impacts of such products would not only be for the environment but also for the health and safety of humans and animals alike (San Paik, 2015).

Green electronics are devices which use as much biodegradable parts so as not to burden the environment excessively. Supercapacitor is one such component which is used in electronic storage devices. Supercapacitors are usually prepared using non-biodegradable materials. However, in recent years researchers are working on supercapacitors fabricated using biodegradable materials. Innovative ventures had led researchers to study polymer electrolytes and the possibilities of making these polymer electrolytes conductive enough to be used in electronic storage devices. Polymers can be both biodegradable and non-biodegradable. In the past two decades, the progress of the portable consumer electronic devices has made an urgent need to develop electrochemical cells with high specific energy and design flexibility. Industries are now pushed to make their products environmentally friendly and sustainable with escalating environmental awareness among consumers. The definition of sustainable has been rewritten to become meeting the needs of the present without compromising the needs of the future (Spiertz, 2010).

#### **1.4 Consumers' Willingness to Pay for 'Green' Products**

Consumers willingness to pay for 'green' products generally means that consumers make trade-offs between product attributes, which includes environmental quality. More often than not, green products are more costly. Hence, there arises the need to investigate whether consumers' are willing to pay the price of going green. Given that the end of production is ultimately consumption, it is equally important to understand consumers' willingness to pay for the green products. This will signal consumers' readiness to purchase and consume these products, hence contributing to the achievement of creating sustainable green economy.

The willingness to pay attribute has been widely studied among consumers, however, the studies have yielded various types of results that each study actually raised more questions than answers. Consumers' have different preferences of green products. Some may opt for organic food and 'green' household items but are not particular about electronics or clothes. Hence, consumers' preference depends on the product and the benefit it holds for them directly. Others may not even be aware of importance of 'green' products.

Most consumer electronics contain harmful toxic devices. To be more specific, they contain potentially toxic metals such as cadmium, copper, lead, nickel and zinc (Force, 2009) and flame retardant substances like polychlorinated dioxins and furans (PCDD/Fs and PBDD/Fs) (Sepúlveda et al., 2010) which have been proved to be harmful to human health (Alaee et al., 2003) and the environment (Petreas & Oros, 2009).



Efforts are being made to implement laws to overcome the negative impact of electronic and electrical waste to the environment. Research is also extensively conducted in finding alternative green materials to be used in making components in these electronic devices.

### **1.5 Research Question**

This study came to be of interest looking into the level of environmental deterioration and the questions about electrical and electronics safety awareness among the general population in Malaysia. We are well aware of the detrimental effects that electronics have on our health and the environment in the long term. This is mainly due to the toxic chemicals used in these electronics. But the questions that arise are using these chemicals the only way to obtain electronic and electrical goods, which are cost – effective and safe as well as not compromised in terms of quality? That is what this study is searching for in the experimental section. By using both biodegradable and non-biodegradable polymer electrolytes, we would like to evaluate the performance of the fabricated electrochemical device. Of course, additives would be needed to improve performance where necessary, but these too have to be environmentally friendly.

Many studies have been conducted on the level of awareness among Malaysians on the environment. Several studies have shown that Malaysian consumers' considered environmentally friendly products when purchasing electrical and electronics for their household in order to protect the environment. This clearly shows that most Malaysians are aware of the negative impact of electrical and electronics to the environment. The questions that comes next is what are they willing to do about it. Are Malaysian consumers willing to purchase green electronics to support the environment cause? And if yes, how much are they willing to pay to go green and what are the determining factors for Malaysian consumers to purchase green electronics. The next thing we wish

to delve further into is how much Malaysian consumers are willing to pay for the green electronics.

The experimental part was carried out first so that it would be able to well complement the survey that will be carried out. The experimental section would be used to explain to consumers' on the existence of green electronics and how it helps in sustaining the environment. This would be done by comparing biodegradable based polymer electrolytes and non-biodegradable based polymer electrolytes which would eventually be fabricated into electrochemical devices. The results obtained would prove that electrochemical devices based on green materials also has better performance when compared with a non-green one, hence the benefit to go green.

This study is novel in few aspects. First, the polymer electrolytes are prepared using biodegradable and non-biodegradable polymers, which contribute to it being green or non-green. Second, a comparative study is carried out to compare the performance of both the polymer electrolytes upon fabrication as a device.

In addition, this study combines experimental and statistical data. A connected survey based on the comparison we have conducted will strengthen the study further and give better understanding to consumers' as it is more evidence based.

## **1.6 Objectives**

1. To prepare and characterize biodegradable and non-biodegradable polymer electrolytes
2. To fabricate electric double layer capacitor (EDLC) using biodegradable and non-biodegradable polymer electrolytes and examine its electrochemical

properties and cyclability for comparison purpose in terms of stability and long-term usage.

3. To investigate the determinants affecting consumers' willingness to pay for green electronics.

## **1.7 Novelty**

There are two (2) types of materials combinations analysed in this work, which is the PVA-  $\text{Mg}(\text{CF}_3\text{SO}_3)_2\text{-BmImBr}$  and PEMA-  $\text{Mg}(\text{CF}_3\text{SO}_3)_2\text{-BmImBr}$ . According to literature, researchers have not studied the combination of polymer-ionic liquid-salt composition reported in this study. In addition, this is a comparative study that compares the performance of biodegradable and non-biodegradable polymer electrolyte compositions by carrying out several characterization methods. As mentioned at the beginning of this chapter, this study goes beyond experiments and ventures into consumers' attitudes and opinions conducted through a survey.

## **1.8 Scope of the thesis**

This thesis would be made up of 7 chapters. The first chapter would be an introduction to this area of work. It would give the reader an overview of what this research's main topics would be, the questions that inspired this research, the objectives of the research and its novelty. Chapter 2 would review related literature related to the topics of research. It would delve into biodegradable and non-biodegradable electronics, the importance of sustainability, green electronics and devices and polymer electrolytes in green electronics. Another important section in this chapter is the analysis of consumers' willingness to pay for green electronics and the criteria for consumers' to go green. Chapter 3 details the materials and methods to carry out this study – both experimental and survey based. The results and discussion would be covered in three chapters, 4,5 and 6. Chapter 4 would discuss the results using biodegradable polymer

electrolytes, Chapter 5 discusses the results using non-biodegradable polymer electrolytes and in Chapter 6 discuss the statistical analysis of the survey conducted on consumers' willingness to pay for 'green' electronics. Chapter 7 would be a comparison and an overall conclusion and summary of the work with ideas for further studies to be conducted.

## CHAPTER 2: LITERATURE REVIEW

### 2.1 Malaysia's Green Future

In December 2009, the Prime Minister of Malaysia made a pledge at the Climate Change Conference in Copenhagen that Malaysia would “*reduce its carbon dioxide emissions to 40% by the year 2020 compared to the levels in 2005, subject to assistance from developed countries*” (Khoo, 2009). The reason for this pledge was because of the continuous rise in carbon emissions around the world whereby in the last few years, developing countries were found to emit five times more carbon compared to the developed countries (NewScientist, 2009). According to statistics, in 2006, Malaysia's carbon emission was 187 million tons or 7.2 tonnes per Malaysian (Khoo, 2009).

After 6 years, in 2014, the Prime Minister attended the climate change conference in New York and announced that Malaysia has successfully reduced its carbon emission intensity of its GDP by 33%. This was achieved through implementation of new national policies on climate change and green technology, passing a Renewable Energy Act establishing a feed-in-tariff for renewables and revisiting the water resource management policies (Bernama, 2014).

Environmental sustainability is now the aim of the Malaysian government and everyone involved including its industries are committed to make Malaysia green. Malaysia's City of the Future or Forest City will be making its debut in the Iskandar region. It boasts as Southeast Asia's first mega mixed-use green development with vertical greenery and smart city design (City, 2016).

When speaking of cities, Malaysia has identified that low carbon cities are the core of Malaysia's green future. Malaysian Green Technology Corporation (GreenTech Malaysia) and Ministry of Energy, Green Technology and Water Malaysia (KeTTHA), will be working with Carbon Trust in identifying ways to ensure the emergence of these low carbon cities in Malaysia under this programme. It is believed that this programme will most importantly ensure expansion of greener and cleaner Malaysian cities which would not only benefit Malaysia but the planet (Rugg, 2014).

When we speak of sustainability, we also need to consider sustainable energy. It is an undeniable fact that day by day we are watching the depletion of energy sources. The world in fact is moving towards an energy revolution towards greener and cleaner energy. Petronas Gas Berhad, one of the world's largest exporters of oil and gas has plans to process LNG from its liquid to gas form and make it available to people for daily use. With the cleanest form of fossil fuel still being gas, they are confident that the country will still grow but in the cleanest and greenest manner (Maierbrugger, 2012)

Further evidence of Malaysia's commitment can be seen through Malaysia participation in the Expo 2017 at Astana Kazakhstan with the theme Future Energy. The expo will be for 3 months and in line with the expo theme, the Malaysian theme will be 'Powering Green Growth'. Malaysia will be "driving the agenda of carbon reduction and climate change with futuristic and green energy technologies as the core drivers, including solar and biomass technologies, energy efficiency practices, electric mobility, low carbon cities, green buildings, smart grids as well as ICT technologies" in the words of the KeTTHA Minister, Datuk Seri Dr Maximus Ongkili (Bernama, 2017). Aside from future energy, the focus would also be on innovations and practical energy

solutions to help Malaysia manage its energy resources and efficiently lessen environmental damage and address energy security (Digest, 2017).

It is an undeniable fact that the greening of Malaysia would have to begin with its people. Likewise, the Malaysian government with the co-operation of Non-Governmental Organizations (NGOs) have employed and implemented many Go Green campaigns to create awareness among the larger public. Well known steps such as “No Plastic Bag Day Campaign”, “3R campaign (Reduce, Reuse, Recycle)” and the very recent “No Plastic Bag in Selangor” campaigns are creating the much wanted awareness among Malaysians (Hosseinpour et al., 2015). But how far are consumers’ willing to go when it comes to going green? Are they willing to spend extra for green products? When it comes to food, Abdul (Abdul, 2009) found that Malaysian consumers who are concerned about health and the environment are more likely to be willing to pay for green products. In Turkey, similar results were obtained where more than 64% respondents had a positive attitude towards the environment and were willing to pay a premium price for organic seafood (Budak et al., 2005). Ward et al found that consumers’ were willing to pay slightly more for refrigerators produced through green manufacturers compared to those that were not (Ward et al., 2011). Generally, there is a positive trend when it comes to consumers going green, like in South Africa where consumers’ were willing to pay a much higher premium for green electricity (Oliver et al., 2011). However, the willingness of consumers’ to be willing to go green has been found to depend on many factors, some of which contradict from one study to another. This will be discussed in later sections.

## **2.2 The ‘Greening’ of Malaysian Electrical and Electronics Industry**

It all began when ISO14000 was implemented and began to give a positive impact on the world. The environmental management systems outlined in the ISO14000 guidelines was in line with good safety standard and practice (Eng Ann et al., 2006). Since then, the subject of going green has risen exponentially and this indeed can be attributed to the awareness created by the campaigns following and also due to the detrimental effect that the world is experiencing (Puvanasvaran et al., 2012).

With the dawn of the National Key Economic Areas (NKEAs), innovation has now become a priority to revitalise the local industry. The electrical and electronics industry has always been a major contributor to the Malaysian economy and hence, it is not surprising that this industry has been in the loop for the NKEA. During the initial phase, emphasis was placed on the industries development enablers to supplement the Malaysian Investment Development Authority (MIDA)’s existing efforts to promote investments and to allow the Electrical & Electronic sector to move towards future developments.

However, upon revisiting the NKEAs goals and with growing competition from the Malaysian neighbours, our electrical and electronics industry is compelled to take greater strides from just being an assembly hub into research and development, design and manufacturing. This includes Solar Module Producers, semiconductors, cost-efficient advanced materials, Solid State Lighting, Electric Vehicle Component Manufacturing and nanotechnology. NKEA also bears in mind the need to go green hence the goals are in line to reinforce the development of green technology in Malaysia (PEMANDU, 2014).



Many companies realize the importance of going green, however they face several difficulties to sustain in the local market which would eventually diminish their opportunities in the global front. A study conducted by Qader (2011) among several electrical and electronic companies in Malaysia showed that many companies comply with most environmental standards and manage their environmental practices. These practices include increasing energy efficiency, reducing waste, reducing the release of toxic emissions and compliance with environmental standards. However, these companies face several losses in the effort to go green. Environmentally friendly materials are expensive, technology is limited and thirdly consumers' go for cheaper products which means many companies are more motivated towards making profit rather than being environmentally friendly.

The adoption of green strategies not only enables the firms to reduce their total costs and risks, but can also help them to increase their revenues and intangible values, such as reputation and trusted brands that might contribute to the firm's competitive advantages (Esty & Winston, 2009; Noor, 2011).

As a developing nation, Malaysia stands in the frontline as one of the countries to spearhead the need for Green Technology. This drive has also been shown by the Prime Minister who has pushed forward this need into mainstream ideas and portfolio. The government is aware that demanding that industries turn to Green Technology is going to increase their cost tremendously and with that in mind, has come up with several incentives to encourage companies to go green. One such scheme is the green technology financing scheme (GTFS) that leads to improve the innovation development in green technology field. Research and development is also undertaken in full swing to support the development of green technology. Innovation and forward thinking is

necessary if Malaysia wishes to take the lead in becoming a Green Technology hub in Asia. However, all these would not be possible if there is no paradigm shift in Malaysian consumers way of thinking towards innovation and green technology. Only then can Malaysia be able to adopt all the opportunities and advantages from that area.

### **2.3 Green Cellphones – Myth or Possibility**

In today's world, cellphones have become a necessity instead of a luxury. This may be due to the fact that the cellphone has now become the primary means of communication. The market for cellphones have reached a near saturation point, but the demand and technology for cellphones have not diminished.

As technology advances, the demand for environmentally friendly electronics are also increasing. The cellphone does not escape this scrutiny. Currently, cellphones contain many different types of toxic chemicals, from the cover to its components. However, with the strict European Union Regulations, Restriction of Hazardous Substances (RoHS), the number of phones being made with cadmium, lead, mercury and other dangerous materials are decreasing.

Many developing nations which are not bound by the EU directive are still using non-RoHS compliant phones. This does not pose a direct health threat. However, once these phones are at the end of their life, they are mostly just thrown away and end up in landfills, causing the toxic substances to seep into the soil. This would then pose as a threat. Growing concern is also for black market phone batteries which are sold for very low prices but with very high levels of mercury. These have been shown to cause birth defects and brain damage. Although the black market trade poses a great threat especially in Asia, many countries in Asia have taken positive initiatives to prevent

illegal e-waste and to improve recycling options so as to reduce health hazards and negative environmental impacts (Leahy, 2006).

In Malaysia, recycling of these electronics are being carried out but the awareness among consumers' is still at a low level, hence, only those with knowledge on electronics recycling participate in the recycling initiatives. In addition, the scope of recyclable items are limited. By 2020, Malaysia is estimated to have a cumulative total of 1.165 billion units or 21.379 million metric tonnes of Waste Electrical and Electronic Equipment (WEEE). Malaysian Motorola and Nokia operated Take Back Programs (TBP) to recycle cellphones, chargers, cables, hands free units and rechargeable batteries. Dell Malaysia accepts Dell and computers of other brands for recycling. It also gives an options to collect old computers from customers in exchange of a purchase of a new Dell computer (UNEP, 2011).

A 'green' cellphone is considered an 'environmental friendly' phone as it would have lesser or no toxic materials and is more energy efficient. The countries that are bound to the RoHS however are on the verge on discovering green cellphones or environmentally friendly cellphones. The new regulations has practically forced the five major cellphone companies to eliminate toxic metals and other related toxic products from their products (Leahy, 2006).

Due to the demands for greener cellphones, several major cellphone companies have come up with environmental friendly or eco-friendly cellphones. In a survey done in 2015, seven cellphones were given the environmental friendly status based on its energy efficiency, use of hazardous and toxic chemicals, use of recycled plastic and of course, energy efficiency. One cellphone was the Microsoft Lumia because it has 0%

plastic, nickel and brominated flame retardents. In addition, it has very low radiation and battery saving options. Sony's list of cellphones aims to achieve zero environmental damage and all their products claim to be environmentally friendly. Samsung has a model which they claim is the greenest of their products due to low GHG emission, low radiation and usage of recyclable materials. Apple is also on the list with their iPhone which has a recyclable enclosure and power adapter with a very good energy saving option. Huawei has produced the Huawei Ascend W1, a cellphone which is good in performance, low hazardous material and made entirely of recycled plastic. Its radiation levels are also deemed to be very low. Currently, the number 1 entirely eco-friendly phone is supposed to be Fairphone, from a Dutch-based company. It is claimed to have exceptional performance and eco-friendly as it is made out of aluminium and recycled plastic. It is considered the phone of the future (Boric, 2015).

A 'green' cellphone is therefore, not a myth. It is becoming a reality if it already has not. What is needed now is awareness on why we should go green and willingness to purchase.

#### **2.4 Biodegradable Materials in the Electrical and Electronics Industry**

The global community has realized that development and economic growth comes with a price and that price has been the deterioration of the environment. Both developed and developing countries have now taken a firm stand towards sustainable environment. With global awareness on environmental friendliness and sustainability, many companies have adopted green technology and have started their manufacturing operations using biodegradable materials or environmentally friendly materials. Green technology is defined as the development and application of products, equipment and

systems used to conserve the natural environment and resources, which minimizes and reduces the negative impact of human activities.

Gas generated hybrid vehicles are now shifting to electric cars as is being done by Toyota. German engineers are researching on jets that are fuelled using hydrogen and not petrol and Daimler-Benz has invested heavily in a Canadian fuel cell venture (Anderson, 2004). On the local soil, the Malaysian Automotive Institute (MAI) through collaboration with the University of Wollongong, University Technology Sydney, AutoCRC Ltd and Swinburne University have developed a prototype of an electric bus and lithium-ion (Li-ion) batteries for electric and hybrid vehicles. Eclimo Sdn. Bhd., a Malaysian company with co-operation from GreenTech, also manufactures electric scooters and lithium-ion battery packs (PEMANDU, 2014).

**Table 2.1: Structure of Electrical and Electronics Industry in Malaysia**

Sectors	Sub-Sectors	Products
Electronics	Components	Semiconductors, passive components, printed circuit boards, metal stamped parts and precision plastic parts
	Consumer	Audio visual products such as television receivers, portable multimedia players (PMP), speakers, cameras and electronic games
	Industrial	Multimedia and information technology products such as computers and computer peripherals, telecommunications equipment and office equipment.
Electrical	Electrical	Boards, panels and consoles, switching apparatus, lamps, air conditioners, vacuum cleaners, ovens, transformers, cables & wires, primary cells & batteries, solar cells and modules

Source: Malaysian Investment Development Authority (MIDA)

Table 2.1 shows the structure of electrical and electronics industries in Malaysia. Wireless technologies, low power consumption microelectronic devices, Internet of Things (IoT), high technology wearables, and environmental sensors are becoming the new sensation in this generation. The Malaysian electrical and electronics industry acknowledges this as can be seen from the table above. These devices are expected to

be ultra-thin, highly flexible, rollable, stretchable, comfortable to the human body, ubiquitous, and almost imperceptible to the users. This is becoming the reason why high-performance electrochemical applications (ECAs) such as batteries, fuel cells, capacitors and solar cells have been in great demand in the past decade. In the race for the invention and development of these high-performance ECAs, the development of the materials is still considered as one of the most challenging tasks to be conquered by the researchers and innovators. The ECAs need to be high in mechanical properties and able to generate high enough electrical properties to power up these high technology devices.

ECAs are normally composed of a combination of components with different functions such as electrolytes and electrodes. Without proper understanding of the materials used for these components, it is very difficult for the researchers to look for the optimum combination to produce high-performance ECAs. In other words, the performance of the ECAs is dictated by the understanding of the limitation of the materials used to produce the components. Not only the types of materials used, but also the state, composition, shape or even sizes (range up to nano) have a significant impact on the performance of the ECAs.

In addition to the urgent need for efficient, clean, and sustainable sources of energy, waste disposal problem and depletion of non-renewable fossil fuel has also catalyzed the development of environmental friendly alternative energy sources. The dilemma of pollution from the plastic waste becomes a main concern in the environmental trepidation because of the lack of degradation after being discarded. Green chemistry deals with the design of chemical products and processes that reduce or eliminate the use and generation of hazardous substances. Utilization of non-toxic

chemicals, environmentally benign solvents and renewable materials are the key issues in the materials science field when green chemistry methods are considered (Brandt & Balducci, 2014; Fletcher et al., 2010). Therefore, a force has been driven to the development of biodegradable materials to replace those non-biodegradable materials. Biodegradable materials have attracted enormous attention worldwide as a result of white pollution, one of the environmental crises. One material that has taken the limelight is polymer.

## **2.5 Brief History of Polymer Electrolytes**

Polymers have always existed naturally in the environment and have played a major role in human existence since the beginning of time. In the 18<sup>th</sup> century, man-made polymers were discovered. In 1907, the first synthetic polymer was invented by Leo Baekeland (Sperling, 2011) which actually was a thermosetting phenol-formaldehyde resin called BAKELITE. In 1920, the German chemist Herman Staudinger (Carraher, 2003; Muelhaupt, 2004) suggested that polymers are giant molecules which are formed through the permanent attachment of smaller molecules. However, he was ridiculed and his work was disputed. It was not until a decade later that his work became accepted and he was bestowed the Nobel Prize in 1953.

The end of World War II saw an unparalleled surge in the world's economic growth. Developments were rapid in the field of chemical sciences but scientific research proved to be too expensive. This rapid economic surge, however, lasted until 1973 when the oil crisis came on (Di Noto et al., 2011). Wright (1998) surmised that the oil crisis marked the end of availability of cheap and abundant energy and resources. Focus was back on scientific research for new innovative devices for energy storage and conversion and this marked the debut of polymer electrolytes. In the early days, polymers were used as structural materials or insulators. However, since 1975, they

have been tailored as ion conductors when combined with the appropriate salts, which enhances their ionic conductivity. This was first discovered by Fenton when his team successfully dissolved alkali salts into poly (ethylene oxide) and obtained a high conductivity and Wright, when he discovered that poly-ethylene oxide (PEO) when incorporated with sodium or potassium salts produces a solid electrical conductor polymer/salt complex (Fenton et al., 1973; Wright, 1975). This discovery was followed by Michel Armand who, using graphite intercalation compounds as electrodes, realized that lithium/PEO complexes could be utilized as solid electrolytes (Armand, 1978). However, the conventional polymer-salt electrolytes exhibit low ionic conductivity and low mechanical strength. From a practical standpoint, at that time, enhancing conductivity of the polymer electrolyte was a challenging feat. Manipulations needed to be employed, and is still being actively conducted, to achieve high conductivity of these polymer electrolytes.

The concept that proved interesting for scientists was the phenomenon of a solid material exhibiting liquid-like conductivity without any solvent motion. In the 1990s, the development of 'lithium-ion' batteries amounts to an exorbitant \$8 billion/year market. However, this battery uses liquid electrolytes which projects a danger to the user and the device it is used for. With the awareness on the dangers using liquid electrolytes, the dawn of a solid electrolyte was welcomed eagerly. Theoretically, it was a fascinating discovery coming at the right time where the world was speaking about pollution and alternative energy.

Research is still rampantly on going with much interest channelled to improving the properties of these solid polymer electrolytes. The scientific community is willing to compromise the liquid electrolytes with the use of solid polymer electrolytes with



acceptable mechanical properties and high conductivities. These “polymer lithium-ion batteries”, in the thin film configuration and plastic electrolyte may one day replace existing energy systems in the electronics market. As the years progress, research is also being conducted on other energy storage systems such as fuel cells, supercapacitors, electrochromic devices and solar cells. The call towards green energy is echoed by the scientific community for the transformation of industries worldwide.

## **2.6 Solid Polymer Electrolytes**

Polymer electrolytes are formed through the complexation that takes place between high molecular weight polar polymers and ionic salts which have low lattice energy and bulky anions (Kim & Kim, 1999). Both the anion and cation in the materials can be mobile (Kawamura et al., 2006). The ionic conduction in polymer electrolytes comes through the covalent bonding between the polymer backbone with ionizing groups. It is the electron donor in the polymer that forms solvation to the cation group in the ionic salt, which then causes ion separation, facilitating the ion hopping mechanism which, in turn, generates ionic conductivity.

Polymer electrolytes take the upper hand when it comes to solid electrolyte systems due to its high mechanical stability, its flexible thin film form which allows good electrode-electrolyte contact for fabrication of electrochemical devices and mouldability. These thin film polymer electrolytes can be prepared using solution casting method, sol-gel method or electro-deposition method. Hot press technique is also practiced (Agrawal & Mahipal, 2011; Karmakar & Ghosh, 2011; Pandey et al., 2008; Thompson et al., 2001).

Good ion conduction behaviour in polymer electrolytes is only observed above the glass transition temperature. Below glass transition temperature, the segmental chain

motion in the polymer electrolytes does not take place and this causes the ionic conductivity to be quite low (Kawamura et al., 2006; Vogel & Torbrügge, 2006).

Polymer electrolytes are divided to four major classes as depicted in Table 2.2 below:

**Table 2.2: Comparison between four major classes of polymer electrolytes (*adapted from Ngai et al., 2016*)**

<b>Types of polymer electrolytes</b>	<b>Gel polymer electrolytes</b>	<b>Solid polymer electrolytes</b>	<b>Composite polymer electrolytes</b>	<b>Liquid crystal polymer electrolytes</b>
<b>Common properties</b>	Light weight; free of solvent; low volatility; safety; high-energy density; good flexibility, easy to process or configure; wide electrochemical window; good chemical, mechanical, photochemical, electrochemical, structural and volumetric stabilities			
<b>Other properties</b>	High ionic conductivity	High automation process, good mechanical strength	High thermal stability; good interfacial contact; high ionic conductivity	Increase carrier properties and electrical conductivity
<b>Advantages/Disadvantages</b>	Poor mechanical strength	Low ionic conductivity at ambient temperature; high interfacial resistance		Improves electrochemical and optical properties of polymer electrolytes

Solid polymer electrolytes (SPEs) are an innovation to replace liquid polymer electrolytes. As efficient as liquid polymer electrolytes are in terms of ionic conductivity and long term use, it posed a safety threat due to its high likeliness of electrolyte leakage (Liu et al., 2017). It was also a threat to the environment. Thus, the scientific world was compelled to turn to innovations like SPEs. The main pulling factor for SPEs was its solid state, which enhances its safety feature.

Over the past decade, solid polymer electrolytes (SPE) have received wide attention mainly due to its numerous applications in batteries, fuel cells, super capacitors and sensors (Park et al., 2006). Ease of processability, lightweight and transparency are the main characteristics of SPEs (Idris et al., 2009).

Solid polymer electrolytes are deemed better than other polymer electrolytes due to the following reasons (Agrawal & Gupta, 1999) :

- Ease of fabrication into thin films – internal resistance can be reduced which would allow an improved current density
- Polymer electrolytes have better safety due the absence of flammable organic solvent
- Polymer electrolytes have better mechanical strength and flexibility
- Polymer electrolytes are generally stable under ambient conditions which facilitated production

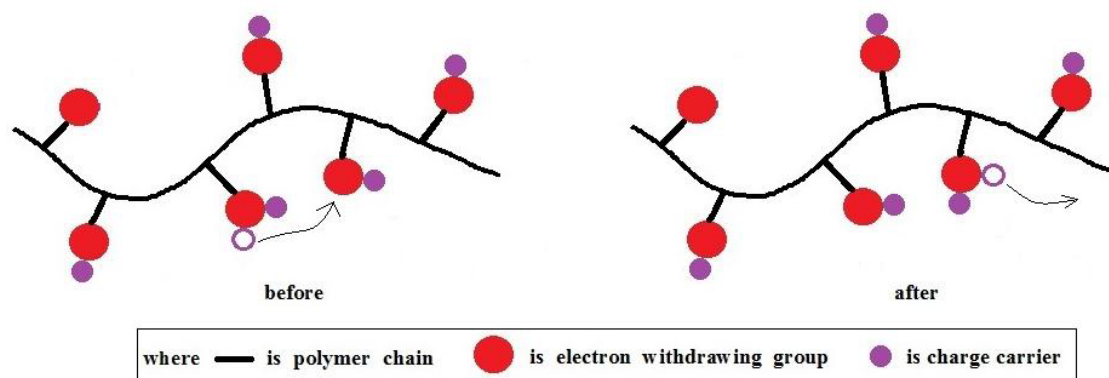
For any material, its potential application would be based on its quality and its efficiency as well as efficacy. Another feature that is stressed now is its safety feature. Safety would not only mean to mankind but also to the environment. Technology should not be compromised to increase the quality and efficiency of a product and at the same time, sustainability should also be taken into consideration to ensure the environment is protected.

## **2.7 Ionic Conductivity of Polymer Electrolytes**

Ionic conductivity can be defined as the movement of ions from one site to another through defects in a solid or aqueous solution. This ionic conduction is usually caused by the hopping of positively or negatively charged ions from one lattice site to

another lattice site under the influence of an electric field. However, for the ions to hop from one site to another, incurring ionic conduction, they would first need to overcome an energy barrier.

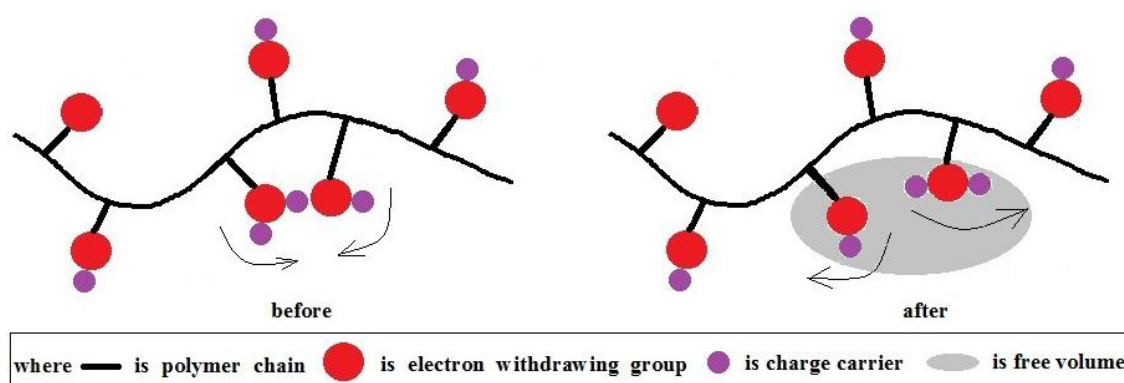
There are two mechanisms for the movement of the ions to incur an ionic conduction in the polymer electrolytes. They occur below and above the glass transition temperatures. The ion hopping mechanism also known as the vacancy mechanism occurs when the ion hops from its original position to an empty adjacent site, hence the name vacancy mechanism. The energy needed for the ions to hop from one site to another empty site comes from the thermal energy of the ionic vibrations which are required to break the coordination bonds to allow ion hopping (Raghavan et al., 2014). When ions migrate from one interstitial position to neighbouring position that is already occupied, an interstitial mechanism would occur (Souquet et al., 2010). That is illustrated in Figure 2.1.



**Figure 2.1: Schematic representation of ion diffusion before and after an interstitial mechanism (Souquet et al., 2010).**

Above the glass transition temperature, a free volume mechanism occurs. This mechanism occurs in addition to the interstitial pair migration mechanism described

earlier and polymer segmental mobility. This mechanism assumes the formation of a ‘cage’. This cage is formed by neighbours closest to the ion during movement of the polymer segments. When there is movement of polymer segments, a free volume will be produced in the polymer matrix, which allows the ion to escape from its cage to an adjoining cage that is large enough to allow an ion jump (Souquet et al., 2010). The illustration for the cage mechanism is depicted in Figure 2.2.



**Figure 2.2: Schematic representation of ion diffusion before and after a free volume mechanism coupled with the chain movement (Souquet et al., 2010).**

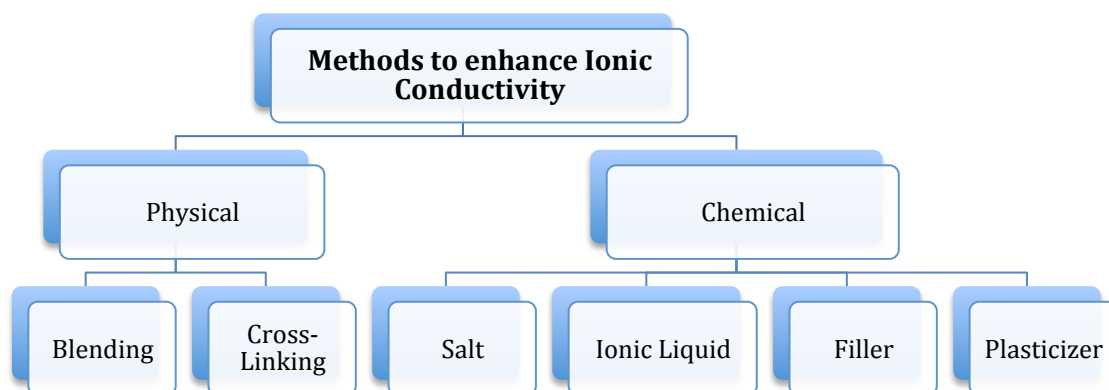
Ionic conductivity of polymer electrolytes can be increased or enhanced by increasing the number of mobile charge carriers and by ensuring that the mobile charge carriers are highly mobile and can be easily detached from its coordination bond. This would depend on factors such as high flexibility of polymer chains, low degree of crystallinity, low glass transition temperature, high amorphous nature of polymer and high concentration of ions.

When polymers are of crystalline nature, their molecules are arranged orderly. In the amorphous state, the molecules or atoms are arranged in a more random order. In an orderly arrangement, it is difficult for ions to be transported or to migrate. Above glass

transition temperature, a crystalline state polymer turns from a glassy state to a soft rubbery state. This state can be considered an amorphous state. The mobility of the ions, which are still restricted in the crystalline state become more mobile above the glass transition temperature in the rubbery state. This transition increases the segmental motion of the ions or molecules along the chains of the polymer and the polymer backbone. The high dielectric constant of the polymers would also favour ion separation and would prevent ion clustering in the polymer electrolyte matrix. The flexibility of the polymer chains also helps in enhancing the ionic conductivity as it increases the mobility of the charge carriers. This is because the ions can be easily dissociated from its interactive bonds in the flexible polymer chains. Ionic conduction hence increases.

## **2.8 Enhancing Ionic Conductivity**

Polymer electrolytes are frequently used to fabricate devices. Successfully obtaining a device with good performance requires a polymer electrolyte with high ionic conductivity. The low ionic conductivity of polymer electrolytes impedes its use in electrochemical devices. Thus, many approaches have been employed to improve the low ionic conductivity of polymer electrolytes to make it usable in electrochemical devices. Figure 2.3 illustrates the methods, which have been employed to enhance ionic conductivity. In this study, salts and ionic liquids will be used to enhance ionic conductivity of polymer electrolytes.



**Figure 2.3: Methods to Enhance Ionic Conductivity (adapted from Ngai et al., 2016)**

### 2.8.1 Salt

Doping with salt has been identified as an effective way to improve conductivity in polymer electrolytes. In addition to the interaction between the polymer host and doping salt, the chemical and mechanical stability of the mixed polymer host and salt also contributes to the improvement in ionic conductivity (Ramesh et al., 2012). By virtue of low lattice energy, salt used as dopant can easily dissociate into cations and anions when in contact with the host polymer which also has low lattice energy but high dielectric constant (He & Kyu, 2016; Ngai et al., 2016). This eventually increases the stability of the polymer electrolyte.

Lithium salt has been the most commonly used as doping salt. Lithium trifluoromethanesulfonate is commonly used as dopant salt compared to other lithium salts due to its resistance to oxidation, thermal stability, nontoxic nature and insensitivity to ambient moisture (Ahmad et al., 2006).  $\text{LiPF}_6$  is used as it has good solubility, low cost and its environmentally friendly feature due to lower hygroscopic properties (Liew & Ramesh, 2014; Ramesh et al., 2011). Lithium bis

(trifluoromethanesulfonyl) imide (LiTFSI) is also a good choice of salt as it has extensive delocalized electrons in TFSI anion which disallows dissociation with Li cations. This in turn encourages lithium transport leading to an increase in ionic conductivity. In addition LiTFSI has also been found to exhibit good thermal and wider electrochemical stability (Ramesh & Liew, 2012). In one study, polymer electrolyte blends incorporated with LiBF displayed slightly higher conductivity compared to polymer films doped with LiClO<sub>4</sub> (Stephan et al., 2000). Other than lithium salts, non-metal salts such as ammonium acetate (CH<sub>3</sub>COONH<sub>4</sub>) are also gaining popularity as dopant salts. The hydrogen bonding in the acetate ion (CH<sub>3</sub>COO<sup>-</sup>) has been found to facilitate salt solvation (Liew et al., 2014b).

There has been no doubt as to the wide usage of lithium in electrochemical applications especially for battery usage. However, the reactivity of lithium metal may have detrimental effect to the environment and users. Thus began the challenge for an alternative and magnesium salt has been found to be a prospective candidate. This will be discussed in later sections.

### **2.8.2 Ionic liquid**

Ionic liquids (ILs), or room temperature ionic liquids (RTILs) are defined as liquids that are entirely composed of ions and are in liquid form at temperatures below 100°C. Ionic liquids are made up of organic salts which, are bulky and asymmetric. Some examples of ionic liquids cations are methylimidazolium [Rmim], N – butylpyridinium [R-N-bupy], N –methyl-N -alkylpyrrolidinium, quaternary ammonium and quaternary phosphonium ions; and anions such as hexafluorophosphate [PF<sub>6</sub>]<sup>-</sup>, alkylsulfates [RSO<sub>4</sub>]<sup>-</sup>, bromide [Br]<sup>-</sup> or iodide [I]<sup>-</sup>, sulfate [SO<sub>4</sub>]<sup>2-</sup>, triflate [CF<sub>3</sub>SO<sub>3</sub>]<sup>-</sup> and bis(trifluoromethylsulfonyl)imide (Brennecke & Maginn, 2001; Holbrey & Seddon,



1999; Meindersma et al., 2010; Wasserscheid & Keim, 2000). As ionic liquid is actually a salt, it is deemed to have negligible vapour pressure which basically means ionic liquids are not volatile. In addition, ionic liquids are miscible with water or organic solvents. Ionic liquids also have high thermal stability. Some ionic liquids are thermally stable till 500°C. Ionic liquids such as 1-ethyl-3-methylimidazolium tetrafluoroborate, 1-butyl-3-methylimidazolium tetrafluoroborate and 1,2-dimethyl-3-propylimidazolium bis(trifluorosulfonyl) imide have been found to remain stable until temperatures of 445°C, 423°C and 457°C, respectively ( Valkenburg et al., 2005). The non-flammable feature of ionic liquids also makes it a prospective candidate to substitute volatile organic compounds (VOC) (Holbrey & Seddon, 1999; Wasserscheid & Keim, 2000).

Studies have shown that ionic liquids have wide electrochemical window, which is a necessary property when ionic liquids are used in electrodeposition of metals and semiconductors. Electrochemical window is defined as the electrochemical potential range within which the electrolyte is neither undergoes reduction or oxidation at an electrode. This value denotes the electrochemical stability of the solvents. In general, the electrochemical window of elements in water is about 1.2.V. Ionic liquids, on the other hand, have higher electrochemical window as studied by x and y who obtained a potential window of 4.15 V for [bmim][PF<sub>6</sub>] at a platinum electrode, 4.10 V for [bmim][BF<sub>4</sub>] respectively (Schröder et al., 2000).

Ionic liquids have also been known to exhibit strong plasticizing abilities. This plasticizing effect helps soften the polymer backbone, which enhances the flexibility of the polymer chain. This phenomenon would lead to easier movement of the mobile charge carrier along the now flexible polymer backbone in the polymer matrix, which

eventually gives rise to higher ionic conductivity. As has been mentioned earlier, a disordered non-crystalline polymer structure would encourage ion mobility. The low viscous nature of ionic liquids assists this by disrupting the ordered polymer electrolyte backbone structure hence decreasing the crystalline region of the polymer backbone (Singh et al., 2009). As there are more free spaces for easier ion migration, ionic conductivity increases. The solubility of ionic liquid is determined by its bulky cations and smaller anions (Jain et al., 2005). The bulky anions which favour delocalization of charges decreases the pairing between cations and anions in low lattice energy polymer complexes (Aravindan et al., 2009). This gives rise to ion detachment of the ionic compound, which undoubtedly increases the ionic conductivity.

## **2.9 Green Polymer Electrolyte**

Synthetic polymers have generally been used as polymer hosts. However, growing concern about clean energy and the detrimental effects of non-renewable energy sources are making scientists turn to nature for natural polymers. Therefore, much effort has been made to develop the biopolymer electrolytes by using natural polymer. Several renewable resource-based biopolymers are suitable to be used as host polymer in the polymer electrolytes, such as starch, cellulose, chitosan, carrageenan, pectin, chitin, lignocellulosic materials, hyaluronic acid, agarose, polylactides, polyhydroxyalkanoates (bacterial polyesters) and soy-based plastics. In addition to being environmentally friendly, these biopolymers could also overcome the main shortcoming of synthetic polymer, which is mostly insoluble in the solvents.

Lignin is an example of biopolymer which is gaining popularity for applications in many fields due to its higher biodegradability and biocompatibility (Gong et al., 2016). Aloe vera is another natural biopolymer that has been blended with PVA to

produce green polymer electrolytes. Aloe vera was used due to the health benefits the plant possesses (Selvalakshmi et al., 2016).

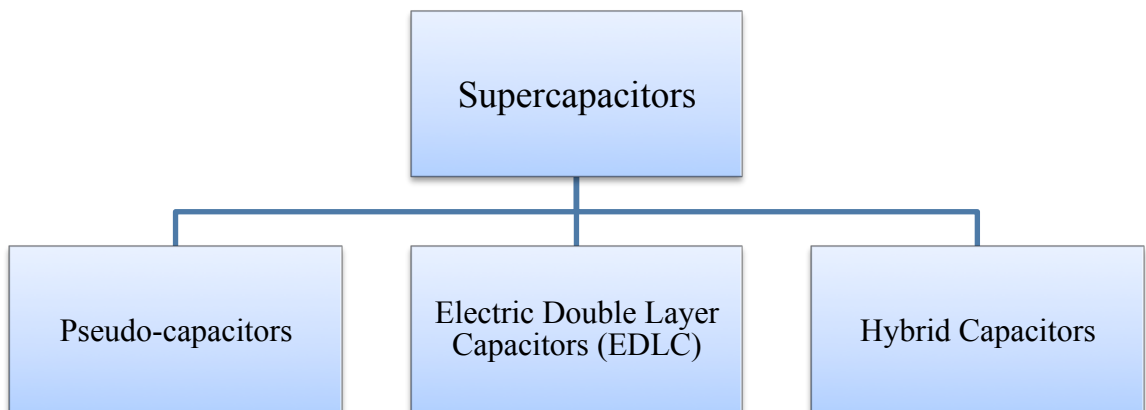
Past research has cited examples such as cellulose acetate (CA) being chosen to build up conducting membranes for use in electrochemical devices. This is due to CA having a number of outstanding properties such as; (a) non-toxic nature, (b) availability of renewable resources, (c) low cost and (d) biodegradable (Avérous et al., 2001; Ramesh & Lu, 2008; Ramesh et al., 2013).

Starch has also gained popularity as a renewable and biodegradable polymer. Starch attracts scientists because of its rich variety and abundances in nature (Kumar et al., 2012). Another natural material Chitosan, is a non-toxic, biodegradable, biocompatible and ability to cast bio films that makes Chitosan to be used in industrial devices (Kadir et al., 2011; Prakash et al, 2013). Chitosan a natural polymer, 1-4 linked 2-amino 2-D glucopyronose, derivative of chitin, which is present in the shells of crustaceans, the cuticles of insects and cell walls of fungi (Sudhakar et al., 2014). The high glass transition temperature and high electrical conductivity at room temperature motivated many scientists to investigate on Chitosan as a polymer electrolyte (Prakash et al., 2013).

## **2.10 Supercapacitors**

Supercapacitors, also known as ultracapacitors, are energy storage devices that can be fully charged and discharged rapidly. Due to the speed, their energy density is lower than batteries but the power it is able to deliver is much higher for shorter durations (Conway, 2013). Thus, supercapacitors are usually used for applications where power bursts are necessary but high energy storage is not. This is due to the

energy storage ability of supercapacitors which is 3-30 times lower than batteries (Miller & Simon, 2008). They can function as complement to batteries or as stand-alone devices. When complemented with batteries, the combination of power and energy improves the energy storage systems, and may improve its lifespan as well (González et al., 2016). However, contrary to batteries, supercapacitors have longer lifespans and cycling abilities without losing their energy storing capacity. Industries use supercapacitors to provide regenerative power whereas for consumer electronics, supercapacitors are made to replace batteries.



**Figure 2.4: Types of Supercapacitors (adapted from Ngai et al., 2016)**

The different types of supercapacitors, as seen in Figure 2.4 can be classified according to its charge storage mechanism or cell configuration. EDLCs energy storage is at its electrode electrolyte interface and is based on high specific-surface area. The electrodes of EDLCs are usually made of carbon-based active materials, which are available abundantly. Pseudo-capacitors, also known as redox capacitors stores energy combining electrostatic and pseudocapacitive charge storage mechanism which relies on fast redox reactions, hence the name redox capacitors. It can hold higher specific capacitance compared to EDLCs. However, the fast redox reaction can lead to

mechanical changes in the electrode (due to the swelling and shrinking of electrodes) making the whole pseudocapacitor mechanically unstable. Hybrid capacitors combine the features of both EDLC electrode and pseudo-capacitive type electrode giving rise to a combination of both systems (Gao & Lian, 2014; González et al., 2016; Simon & Gogotsi, 2008).

### **2.10.1 Electric Double Layer Capacitors (EDLC)**

Early description of EDLC was that it stores charge electrostatically using reverse adsorption of ions. The ions, which are from the electrolyte, were believed to adsorb onto active electrostatically stable materials with high surface area. When polarization occurred at the electrode electrolyte interface, charge separation takes place which gives rise to the double layer capacitance. Researchers later suggested the idea of a presence of a diffuse layer in the electrode due to ion accumulation near the electrode surface (Pandolfo & Hollenkamp, 2006). The advantage of this surface storage mechanism is that energy uptake and delivery is rapid, improving performance. The disadvantage is that, as EDLCs follow the surface charging mechanism, the device would have compromised energy density (Simon & Gogotsi, 2008). Applications for EDLCs now include transportation, aerospace systems, and electronics (Miller & Burke, 2008). Reports have stated the use of EDLCs in emergency exits of an A380 airplane (Rutronik, 2015). This was accomplished by using a parallel assembly of 100-F, 2.7-V cells which are directly incorporated in the doors. This prevents use of heavy copper cables.

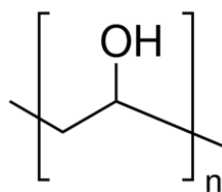
Recently, solid polymer electrolytes have been discovered as potential candidates to be applied in EDLC (Lim et al., 2014). These solid polymer electrolytes can overcome the drawbacks of liquid electrolytes such as reduced reactivity and leakage of electrolyte, improved safety, better shape flexibility and better manufacturing

integrity. These improvements can then enhance the performance of the EDLCs (Pandey et al., 2011). Pandey et al. prepared the polymer electrolytes using polyethylene oxide with inclusion of ionic liquid and upon fabrication of EDLC obtained a specific capacitance of 1.7 - 2.1 F g<sup>-1</sup> in the lithium-based system (Pandey et al., 2011). EDLC based on amorphous SiO<sub>2</sub> nanofiller and poly(ethylene oxide) (PEO)/1-ethyl-3-methylimidazolium hydrogensulfate (EMIHSO<sub>4</sub>) electrolyte were prepared by Ketabi and Lian (2013). The EDLC using this polymer electrolyte exhibited the specific capacitance of 2 mF cm<sup>-2</sup>. Lim et al. (2014) fabricated EDLCs based on activated carbon electrodes and poly (vinyl alcohol) - lithium perchlorate (PVA-LiClO<sub>4</sub>)-nanosized titania (TiO<sub>2</sub>) doped polymer electrolyte found that upon incorporation of 8% nanosized titania obtained a specific capacitance of 7.3-12.5 F g<sup>-1</sup>. Liew et al. (2015) carried out EDLC studies with poly(vinyl alcohol) (PVA)/ammonium acetate (CH<sub>3</sub>COONH<sub>4</sub>)/1-butyl-3-methylimidazolium bromide (BmImBr) and obtained a specific capacitance of 21.89 Fg<sup>-1</sup> with 60 % BmImBr. Another study by the same author using biopolymer electrolytes (corn starch, lithium hexafluorophosphate (LiPF<sub>6</sub>) and ionic liquid, 1-butyl-3-methylimidazolium hexafluorophosphate (BmImPF<sub>6</sub>)) as green polymers for EDLCs exhibited specific capacitance of 37.07 Fg<sup>-1</sup> (Liew & Ramesh, 2015). Developments are continuously being made to obtain improved polymer films for use in EDLCs.

## **2.11 Selection of Materials for the Study**

### **2.11.1 Poly (vinyl alcohol)**

Poly (vinyl alcohol) or PVA is a polymer with carbon chain backbone having hydroxyl groups attached to its methane carbons. The accepted structure of PVA is given in Figure 2.5.



**Figure 2.5: Chemical Structure of PVA**

The hydroxyl groups of its carbon chain backbone is attached to methane carbons, whereby these O–H groups can be a source of hydrogen bonding and therefore, assist the formation of polymer complexes (Hirankumar et al, 2004; Rajendran et al., 2004; Saroj & Singh, 2011). PVA has also been reported to have high tensile strength, in addition to abrasion resistance, which facilitates its use as binder, and in other electrochemical devices (Coleman & Painter, 1995; Rajendran et al., 2004).

PVA is a good material to be used as host polymer for biodegradable polymer electrolytes. It has high glass transition temperature ( $T_g$ ) (358 K) and good mechanical properties (Saroj & Singh, 2012). It also has good charge storage capacity, due to being a polar polymer, high dielectric strength and dopant-dependent electrical and optical properties. Above all that, PVA has excellent mechanical property and shows both ionic and electronic conduction (Rajendran & Mahendran, 2001).

Literatures on PVA studies provide knowledge on the improvement in ionic conductivity of PVA polymer electrolytes. Jeong et al (2006) showed the effect of addition of salt on ionic conductivity with a variation of salt content and dimethyl sulfoxide (DMSO) as solvent and obtained highest ionic conductivity of  $5.41 \times 10^{-4} \text{ Scm}^{-1}$ . Rajendran and co-workers carried out a study on PVA/PMMA blends with different Li-salt contents and the highest conductivity obtained was in the order  $10^{-3} \text{ Scm}^{-1}$  (Rajendran et al, 2003). PVA based system complexed with lithium salt having

relatively high conductivity of the order of  $10^{-8} - 10^{-4} \text{ Scm}^{-1}$  have been reported previously (Liew et al., 2014a; Liew et al., 2015; Lim et al., 2014). Another study has reported that PVA – lithium triflate system has produced ionic conductivity between  $10^{-8}$  to  $10^{-4} \text{ Scm}^{-1}$  (Every et al., 1998). These studies report a polymer electrolyte-salt system. Newer studies are reporting on the effect of ionic liquid on ionic conductivity. Thanganathan and co-worker published a study on PVA based hybrid membranes with phosphomolybdic acid (PMA), 1-butyl-3-methylimidazolium Bis (trifluoromethanesulfonyl) imide (BMITFSI) and 1-ethyl-3-methylimidazolium tetrafluoroborate (EMI-BF<sub>4</sub>) ionic liquids with SiO<sub>2</sub> inorganic filler. A maximum ionic conductivity of  $0.83 \times 10^{-3} \text{ Scm}^{-1}$  and  $0.58 \times 10^{-3} \text{ Scm}^{-1}$  was obtained for the PVA/PMA/SiO<sub>2</sub>/BMITFSI and PVA/PMA/SiO<sub>2</sub>/EMI-BF<sub>4</sub> hybrid composite membranes, respectively (Thanganathan & Nogami, 2015).

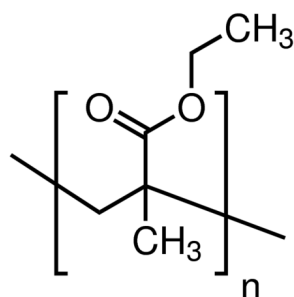
To the best of our knowledge, magnesium triflate based PVA with BmImBr as an additive has not been reported. Although PVA blended with ionic liquid has been reported, the combination of polymer, salt and ionic liquid is novel as this study aims to study and compare the effect of additives in biodegradable and non-biodegradable polymer electrolytes.

Other features of PVA which makes it highly favourable is that PVA is non-toxic, has excellent adhesive and film forming properties, and resistant to solvents. PVA is also fully degradable and dissolves easily. This nature makes it the polymer of choice when preparing biodegradable polymers for green electrochemical devices. PVA has a melting point of 230°C when fully hydrolyzed and a melting point of 180° – 190°C in partially hydrolyzed grades. In addition, its decomposition occurs rapidly above 200°C due to its ability to undergo pyrolysis (Fromageau et al., 2003; E. M. Sheha et al., 2015).



### 2.11.2 Poly (ethyl methacrylate) (PEMA)

Poly (ethyl methacrylate) or PEMA is a derivation of the methyl acrylate polymers and has often been used as host polymer due to their large pendant group structure. Ionic conductivity of polymer electrolyte containing PEMA has been reported in the order of  $10^{-3} \text{ Scm}^{-1}$  (Han et al., 2002). For the development of high conducting polymer electrolytes, a flexible backbone is a mandatory characteristic (Ramesh et al, 2014). With the necessary manipulation, such as the addition of salts or plasticizers, PEMA can be attuned to fulfil the requirement as a high performing polymer electrolyte. The structure of PEMA is as depicted in Figure 2.6.



**Figure 2.6: Chemical structure of PEMA**

PEMA, a methacrylic ester polymer, has also been proven to have excellent ionic conductivity, chemical resistance and high surface resistance (Reiter et al., 2009). In addition, PEMA is also an amorphous polymer that can be used in both optical and non-optical devices (Costache et al., 2006). Rajendran et al. (2013) showed in a study that PEMA added with LiClO<sub>4</sub> and EC showed a highest ionic conductivity of  $2.0245 \times 10^{-5} \text{ Scm}^{-1}$ .

Han and co-workers first carried out conductivity measurement of polymer electrolytes based on PVC/PEMA blends as well as mechanical and morphology studies (Han et al., 2002). They found that the mechanical strength of PVC/PEMA was better

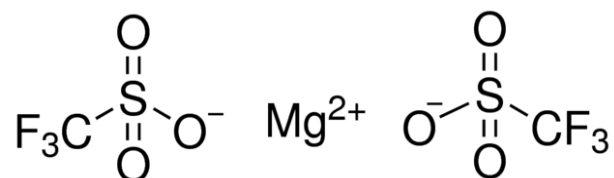
than that of PVC/PMMA based electrolytes. The elongation strength of PEMA was found to be 7% higher and its  $T_g$  (336K) lower when compared to PMMA. Rajendran et al. (2008) showed that PEMA recorded a maximum ionic conductivity of  $6.726 \times 10^{-3} \text{ Scm}^{-1}$  at 303 K with PVC/PEMA blends. In another study with sodium ion conducting PEMA/PVC blend polymer electrolyte, ionic conductivity of  $5.01 \times 10^{-9} \text{ Scm}^{-1}$  was reported (Mohan et al., 2011). Later studies have shown that PEMA as a host polymer has ionic conductivity in the  $10^{-3} \text{ Scm}^{-1}$  order and electrochemical stability of 4.3 V which becomes very useful in solid-state battery fabrication (Rajendran et al., 2008; Zakaria et al., 2012). Vivek and co-workers have experimented with PEMA for dye-sensitized solar cells and obtained an ionic conductivity of  $1.80 \times 10^{-5} \text{ Scm}^{-1}$  with 60 wt% concentration of  $\text{NH}_4\text{I}$  salt (Singh et al., 2015). Sim et al. (2016) reported the ionic conductivity of PEMA/PVdF–HFP:LiTf improved from the order of  $10^{-7} \text{ Scm}^{-1}$  to  $10^{-5} \text{ Scm}^{-1}$  with the addition of 12.5wt.% BMII.

In this study, PEMA represents the non-biodegradable polymer electrolyte, whose efficiency would be compared with the PVA based polymer electrolyte. As with PVA, the combination of polymer, salt and ionic liquid is novel, as this combination has not been reported previously, to the best of our knowledge. This study will compare the efficiency of the biodegradable polymer electrolyte with a non-biodegradable polymer electrolyte and its reliability upon device fabrication.

### **2.11.3 Magnesium Trifluoromethanesulfonate ( $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ )**

Magnesium salt is an alternative to lithium salt as dopant. Magnesium is a divalent salt and has stronger coulombic interactions which would have structural implications on the formation of solid polymer electrolytes (Ramesh et al., 2014). According to the same authors, higher content of salt would have a negative effect on

the ionic conductivity. However, due to lower cost and being a less reactive metal, magnesium is still preferred (Ramalingaiah et al., 1996). The structure of  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  is illustrated in Figure 2.7.



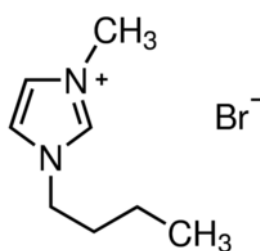
**Figure 2.7: Chemical structure of  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$**

Magnesium is indeed the most abundant element on earth and it also has relatively higher stability. This also makes it a favoured candidate for electrolyte materials. Chusid et al. (2003) mentioned that magnesium systems in electrochemical research is as favoured as lithium based systems as both systems are similar in electrochemical properties. Lithium rechargeable batteries are well known in the electronic world, as opposed to magnesium rechargeable batteries. Yoshimoto et al. (2003) researched the development of magnesium in a non-aqueous media for the development of rechargeable batteries. Following Yoshimoto's study, other studies have also showed that rechargeable magnesium batteries are more preferred due to their high stability with high negative standard potential (-2.375 V), high melting point (649°C), being free from hazards making it highly safe and easy to handle, low cost, relatively low equivalent weight (12g per Faraday) and low toxicity which facilitates waste disposal (Kumar & Munichandraiah, 2001; Sheha & El-Mansy, 2008). Magnesium cells have been successfully produced using poly(vinylidene fluoride) (PVDF) and poly(methylmethacrylate) (PMMA) as polymer hosts and manganese oxide ( $\text{MnO}_2$ ) as cathode (Kumar & Munichandraiah, 2001 & 2002). Zainol et al incorporated magnesium as salt in a PMMA polymer matrix and obtained a high ionic conductivity

of  $1.27 \times 10^{-3} \text{ Scm}^{-1}$  when 20 wt.% of  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  salt was added (Zainol et al., 2013).

#### 2.11.4 1-butyl-3-methylimidazolium Bromide (BmImBr)

The ionic liquid used in this study is halide –based with the chemical structure as depicted in Figure 2.8. This ionic liquid carries the name 1-butyl-3-methylimidazolium Bromide.



**Figure 2.8 :Chemical Structure of 1-butyl-3-methylimidazolium Bromide.**

BmImBr has been discovered to be miscible in water, methanol and dichloromethane but not miscible in acetone, toluene, ethyl acetate and diethyl ether. The cation  $\text{BmIm}^+$  is easily detached from its coordinative bonds with counteranion in the ionic liquids. This detachment is due to the bulky size of the cation which also facilitates ion migration in polymer electrolytes. These ionic liquids are also favoured because of their ability to be recycled and solvation ability at room temperature, which makes them favoured as green solvents (Zare et al., 2011).

The most common problem in solid polymer electrolytes is their low conductivity, which impedes their usage in electrochemical devices. We also want to study how much the addition of ionic liquid, 1-butyl-3-methylimidazolium bromide (BmImBr), would improve ionic conductivity of both biodegradable, PVA– $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  and non-biodegradable PEMA– $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  polymer electrolyte

systems. We go one step further in preparing Electrical Double Layer Capacitors (EDLC) using the highest conducting polymer electrolyte from both systems.

## **2.12 Consumers' Willingness to Pay for Green Electronics**

Electrical and electronics equipment have become a necessity in our everyday lives. Modern technology has brought forward a myriad of equipment to make life easier for us, however that same technology failed to give us a solution for these electronic waste.

This survey is a follow up on the experimental work done on this project. The objective of the study is to investigate the usage of biodegradable and non-biodegradable components in an electronic device making the biodegradable device more environmentally friendly. The device fabricated was an electric double layer capacitor (EDLC), a type of supercapacitors. Supercapacitors are basically energy storage components. Of recent years supercapacitors have gained popularity due to its fast charging and energy storage abilities.

Most consumer electronics contain harmful toxic devices. To be more specific, they contain potentially toxic metals such as cadmium, copper, lead, nickel and zinc (Schluep et al., 2009) and flame retardants like polychlorinated dioxins and furans (PCDD/Fs and PBDD/Fs) (Sepúlveda et al., 2010) which have been proved to be harmful to human health (Alaee et al., 2003) and the environment (Petreas & Oros, 2009). To overcome the negative impact of electronic and electrical waste to the environment, the EU implemented the waste electrical and electronic equipment (WEEE) directive in August 2005, which advises on reusing and recycling electrical and electronic equipment.

To further negate the damage caused by the electric and electronics industry, the RoHS (Restriction of Hazardous Substances) directive was enacted, prohibiting electrical and electronics containing toxic substances such as lead, mercury, cadmium, chromium, polybrominated biphenyls (PBB) and polybrominated diphenylethers (PBDE).

When a product is environmentally friendly and has high safety features, naturally it would be more highly priced compared to their conventional counterparts. Many studies have been conducted on consumers' willingness to pay for green products both internationally and locally, for example, Saphores et al found that a high number of Californian households are willing to pay between 1% and 5% higher for "green" electronics. The study also concluded that the wealthier households were willing to pay a premium of 5% and 10% higher for "green" electronics (Saphores et al., 2007; Saphores et al., 2012). This low willingness to pay may be caused by lack of awareness on the toxicity of the materials used in electronics or total ignorance on the effect these toxic materials have on the environment. Another study found that US consumers are willing to pay average between USD 8.53 and USD 24.21 for green cellphones (Milovantseva, 2013).

On the local front, Ong et al. (2015) found that Malaysian consumers were willing to pay higher for green air conditioners and light bulbs compared to the conventional non-green featured electronics. However, the price they were willing to pay was still lower than the market price of the green electronics available in the market. In a study conducted by Hassan & Nor (2013) on consumers' behavior towards green electronics, the researchers concluded that the consumers' decision making on purchasing green electronics is still weak. This could be attributed once again to lack of

knowledge on existence of green electronics or the effect of the hazardous materials in these electronics.

## **2.13 Factors Affecting Consumers' Willingness to Pay for Green Electronics**

Many factors have been attributed to influencing consumers' willingness to pay for green electronics. In this study, consumers' willingness to pay is assumed to be a function of the below variables:

### **2.13.1 Environmental concerns**

Grob proposed that the more conscious/aware consumers are of the environment, the more they will act in an environmentally conscious manner (Grob, 1995; Maloney & Ward, 1973). Consumers environmental concerns are mainly related to their interest towards the environment and the consumer's surroundings (Suki, 2013). In a study by Afroz et al. (2013), 65% Malaysian households stated that they considered environmentally friendly products when purchasing electrical and electronics for their household in order to protect the environment. This clearly shows that most Malaysians are aware of the negative impact of electrical and electronics to the environment.

Healthy communities begin with environmental concern and environmental friendly attitudes. This is because the decisions we make with regards to the environment now would influence the health and quality of life of our future generations (Royne et al., 2011).

Abdul-Muhmin (2007) stated that environmental psychology could explain the consumers' willingness to be environmentally friendly. Concern towards the environment can be assumed to be the key determinant of willingness to pay.

Conclusively, environmental concerns can be concluded as a perceptual factor, which basically represents a sense of environmental responsibility based on the probability of environment deterioration due to consumers behavioural pattern or belief (Abdul - Muhmin, 2007; Biswas & Roy, 2016). Hence, in our study this variable is expected to have a positive relationship with willingness to pay because more and more Malaysians are becoming environmentally aware and concerned. This concern is not only for the environment but also indirectly for the consumers' own quality of life as well.

### **2.13.2 Health and safety aspects**

When it comes to food, the general assumption is that consumers would go organic due to it being healthier compared to normally grown food. In a study by Bonny (Bonny, 2006), health and safety is found to be the highest motivating factors for consumers to purchase organic food. In the same study 26% stated environmental factors as motivation to go organic. One main reason is due to increase in awareness about health and food safety from usage of synthetic fertilizers and pesticides and concerns about biotechnology in agriculture (genetically modified organisms). Previous studies have shown that health and safety considerations affected consumers purchasing decisions (Gracia & de Magistris, 2008; Hamzaoui Essoussi & Zahaf, 2008).

This result supports many researchers claim that safety and health are currently prominent factors in determining people's attitudes towards the environment. This



assumption supports the idea that the motivation for this behaviour is supported by perceptions of personal gain, which in this case is improved health.

Health consciousness is an important psychographic variable in further developing audience segmentation pertaining to health issues. Midmore et al. (2005) stated that health consciousness can be categorised into two parts; firstly is the product characteristic towards the freshness, naturalness and etc; secondly is the awareness by themselves due to health problem in which organic food is needed to control and maintain their health. In these studies, health and safety has always contributed towards consumers decision making to go green. Likewise, in our study, health and safety is also expected to play a role in determining consumers' willingness to pay for green electronics. However, the question arises as to how much they are willing to pay to maintain their safety and health and at the same time protect the environment. As the study population would be made up of educated working group, we can hypothesize that there may be a positive strong positive link between health and safety and willingness to pay for green electronics.

### **2.13.3 Peer pressure / Social value**

Consumers who purchase green products basically not only help to preserve and protect the environment, they also motivate other consumers to do so (Sheth, Newman, & Gross, 1991). This is because the others would wish to emulate their friends' or relatives' behaviour. This is especially true if the person buying green is part of a group or social circle. However, Biswas and Roy (2015) have reported that there is an insignificant link between social influence and green product purchase behaviour and consumer choice behaviour (Lin & Huang, 2012).

Some consumers tend to feel bad should they not buy green products. They feel that they are behaving responsibly when they make green purchasing decisions (Lin & Huang, 2012). In a study by Bei and Simpson (1995), 89.1% of consumers felt that they were saving the environment when they purchased recycled green products. They think of themselves to be environmentally conscious people as they are helping to save the earth by going green.

Based on previous studies, we believe that in this study, a positive relationship would be obtained between social value and willingness to pay. This is because, Malaysians are people who like to blend in with the trend. Going green to save the environment is a topic discussed in many forums and there are many NGOs which are lobbying to go green. Hence, based on our population, which would be made up of working professionals, who feel the need to adhere to the trend of the day, a positive influence can be expected between peer pressure and willingness to pay.

#### **2.13.4 Price awareness/Availability**

The definition of 'green product' is something that is non-toxic, originally grown, doesn't pollute the environment, contains natural ingredients, recycled content and contains only approved chemicals, if any (Mishra & Sharma, 2010). Most consumers would attribute their willingness to pay for a product on its price, especially if the claimed green product is more expensive (D'Souza et al., 2006). However, past studies found that environmentally conscious consumers from Thailand, Malaysia and Korea were willing to pay premium price for environmentally friendly products (Lung, 2010). D'Souza et al. (2006) noted that products should be environmentally safe and consumers should not need to pay a higher price just because a certain product is environmentally friendly. This was also speculated by Saphores et al. (2012) in a study

on green electronics whereby consumers probably expect manufacturers to be more innovative in producing green electronics without significantly increasing the price as this is the one way to ensure continuous progress in electronics manufacturing.

For this study, it is conjectured that price awareness would highly contribute to consumers' willingness to pay for green electronics. The hypothesis we put forth is that consumers' price awareness significantly influences their willingness to pay for green electronics.

#### **2.13.5 Knowledge/Information available**

A certain amount of knowledge about green electronics and its benefits would influence consumers' willingness to pay for the products. Aryal reported that knowledge about organically produced foods will affect consumer purchase decision (Aryal et al., 2009). Knowledge has direct and indirect effects on attitudes toward consumer to choose the products, and the willingness to pay a price premium. Thus knowledge is the most important factor that determined the demand of organic food.

Product knowledge is an important tool to differentiate the feature and quality of products (Aryal et al., 2009). According to Pieniak et al. (2010), the level of product knowledge can be categories into three sections which are consumer's subjective knowledge, objective knowledge, and also general attitudes. Subjective knowledge can be considered as an important factor as it is a personal knowledge that is interpreted by our personal experiences whereas objective knowledge is referring to the knowledge that gets from the actual information.

This can also be linked to brand awareness (Suki, 2013). Consumers' would be less confident in purchasing a product, even if deemed green, if they are unfamiliar with its brand name. If a brand is associated with green products, there is a higher likelihood of consumers' purchasing from that manufacturer. Suki found that information available on the product and the credibility of the information plays a significant role in consumers' decision on purchasing green products.

In today's world, social media is depended to carry information and news faster than mass media. The influence of social media is infinite. Taking into consideration the state of the world today and the rate at which information is being disseminated, we believe that knowledge and information on a product would have a positive impact on consumers' willingness to pay as well.

#### **2.14 Summary**

In summary, this chapter discussed in detail on the available current literature with regards to biodegradable and non-biodegradable polymer electrolytes, electric double layer capacitors, factors influencing ionic conductivity of polymer electrolytes, ways to improve ionic conductivity of polymer electrolytes and on the green industries in Malaysia. In addition, the factors that have been reported to affect consumers' willingness to pay for green products have also been discussed.

## CHAPTER 3: MATERIALS AND METHODS

This chapter would be divided to two main sections. The first section would address the experimental part of the study where lab experiments were carried out. The materials used for the research and the methods for sample preparation and characterization have been reported. The self-prepared electrode preparation would be in the following part and the last part of the experimental section would be on the fabrication and characterization of EDLC.

The second section of this chapter would explain about the survey part of the study. The survey is linked to the experimental section with relation to the fact that we will be investigating consumers' willingness to pay for green cellphones, which is conducted in the experimental part of the study – the use of biodegradable polymer electrolytes in EDLCs. The theoretical framework of the study, which will include all the variables used, will be discussed. Furthermore, the methodology used for the survey and the rationale behind that will be explained. It would also explain about the sampling procedure, the target samples and the analysis method used to obtain conclusions for the survey. Finally the limitations of the used survey method would be presented and followed by the hypothesis we adopted to test in the survey.

### 3.1 Materials

Table 3.1 shows the materials used in this research and their respective functions. All the materials were used as received without further purification.

**Table 3.1 : Materials used in Study**

<b>Function</b>	<b>Material</b>	<b>Procured from</b>
Host Polymer	Poly (vinyl alcohol) (PVA) – MW 130000 g mol <sup>-1</sup>	Sigma–Aldrich, USA
	Poly(ethyl methacrylate) (PEMA) – MW 515000 g mol <sup>-1</sup>	Sigma–Aldrich, USA
Salt	Magnesium triflate (MgCF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	Sigma–Aldrich, USA
Ionic Liquid	1–butyl–3–methylimidazolium bromide (BmImBr)	Merck, Germany
Solvent	Acetone	Merck, Germany
	Distilled water	-

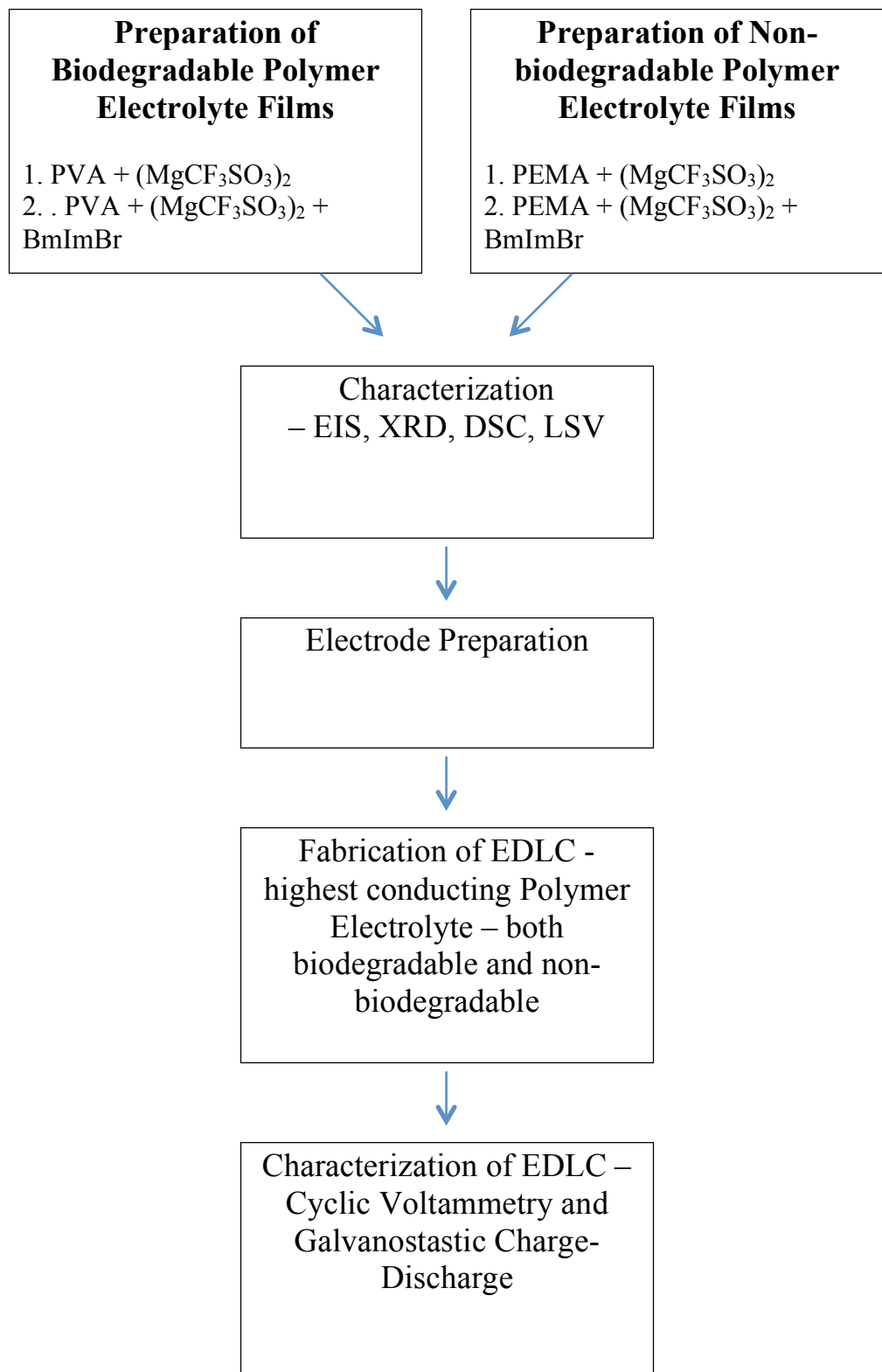
### 3.2 Research Layout

Before the research was carried out, much background work was done on deciding the best polymers to be used, as one of the objectives of our study was to compare between biodegradable and non-biodegradable polymers.

Upon deciding the polymers to be used, salt, magnesium triflate was first incorporated into the polymers and films were formed. The miscibility of the salt and polymer gave us more confidence to proceed to the adding of the selected ionic liquid, BmImBr. These formed films were subjected to ionic conductivity analysis so that it could be decided which composition had the highest ionic conductivity. The polymer electrolytes films were then subjected to thermal, structural and electrochemical characterization. Upon completion of characterization, a device was fabricated using the

highest conducting polymer electrolyte. It was important to use the highest conducting polymer electrolyte, as it is believed that the highest conducting polymer electrolyte would also produce a highly efficient device. The device was also subjected to analysis to investigate its performance.

Figure 3.1 depicts a flowchart of the process followed, from the preparation of the polymer electrolyte films to the fabrication and characterization of the device.



**Figure 3.1: Flowchart of the prepared system of the polymer electrolytes**



### 3.3 Preparation of PVA and PEMA based Polymer Electrolytes

Ionic liquid incorporated PVA based polymer electrolytes were prepared using the solution casting method. The PVA was first dissolved in distilled water. Magnesium triflate was then added into the polymer solution at a ratio maintained at 70:30. Subsequently, BmImBr was added into the solution and stirred thoroughly at 70 °C for a few hours to ensure that a homogenous mixture is obtained. The final step was to pour the mixture into Teflon coated glass Petri dish and dried in an oven at 60 °C to obtain free-standing polymer electrolyte films. The ratio calculation and designation given to the ionic liquid added polymer electrolytes are as in Table 3.2.

**Table 3.2: The weight ratio of PVA, Mg(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub> and BmImBr and their designations**

Designation	Materials (wt.%)		
	PVA	Mg(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	BmImBr
PVA 0	70 (0.7g)	30 (0.3g)	0
PVA 1	63 (0.63g)	27 (0.27g)	10 (0.1g)
PVA 2	56 (0.56g)	24 (0.24g)	20 (0.2g)
PVA 3	49 (0.49g)	21 (0.21g)	30 (0.3g)
PVA 4	42 (0.42g)	18 (0.18g)	40 (0.4g)
PVA 5	35 (0.35g)	15 (0.15g)	50 (0.5g)
PVA 6	28 (0.28g)	12 (0.12g)	60 (0.6g)
PVA 7	21 (0.21g)	9 (0.09g)	70 (0.7g)

For the preparation of PEMA based polymer electrolytes, the same solution casting method was used. However, PEMA,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  and BmImBr were dissolved in 20 ml acetone in the composition as in Table 3.3 to prepare the ionic liquid doped polymer electrolytes. The resulting solution was stirred thoroughly for several hours to ensure that a homogenous mixture was obtained. The mixture was then poured onto Teflon coated glass Petri dish in a fume hood and allowed to set at room temperature. This eventually produced free-standing polymer electrolyte films.

**Table 3.3: The weight ratio of PEMA,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  and BmImBr and their designations**

Designation	Materials (wt.%)		
	PEMA	$\text{Mg}(\text{CF}_3\text{SO}_3)_2$	BmImBr
<b>PEMA 0</b>	70 (0.7g)	30 (0.3g)	0
<b>PEMA 1</b>	63 (0.63g)	27 (0.27g)	10 (0.1g)
<b>PEMA 2</b>	56 (0.56g)	24 (0.24g)	20 (0.2g)
<b>PEMA 3</b>	49 (0.49g)	21 (0.21g)	30 (0.3g)
<b>PEMA 4</b>	42 (0.42g)	18 (0.18g)	40 (0.4g)

The prepared PVA and PEMA based polymer electrolyte films are kept in a desiccator to prevent any traces of water or further absorption of water from the atmosphere. Both the biodegradable and non-biodegradable polymer electrolyte films were further analysed using different characterization techniques as will be discussed in the following parts.

### 3.4 Characterization of Ionic Liquid Added Polymer Electrolyte Film

The characterization methods used to analyse the polymer electrolyte films will be discussed in the following sections.

#### 3.4.1 Electrochemical Impedance Spectroscopy (EIS)

Impedance spectroscopy is a measurement of real and imaginary parts of impedance at different frequencies. The motive of conducting this measurement is to separate the true bulk conductivity from conduction due to reasons such as electrode-electrolyte interface and grain boundary. Electrochemical impedance spectroscopy (EIS) has been widely used and has gained recognition as an important tool in the study of electrochemical devices, corrosion and electro-organic synthesis.

In impedance spectroscopy, when a monochromatic signal  $V(t) = V_m \sin(\omega t)$  is applied across a sample a current  $I(t) = I_m \sin(\omega t + \theta)$  is measured. Here,

$$\omega = 2\pi f$$

$$\theta = \text{phase difference between voltage and current}$$

$$V_m = \text{the maximum voltage}$$

$$I_m = \text{the maximum current}$$

$\theta$  is zero for purely resistive behaviour. The conventional impedance is defined as,  $Z(\omega) = V(t) / I(t)$ . Its magnitude or modulus is  $|Z(\omega)| = V_m / I_m(\omega)$  and phase angle is  $\theta(\omega)$ . The  $\theta$ ,  $V_m$  and  $I_m$  change with frequency, hence, a set of complex impedance,  $Z$ , values are acquired. From the values, a set of real impedance,  $Z_R$  and a set of imaginary impedance,  $Z_I$  are then calculated.

$$Z_R = Z' = |Z(\omega)| \cos \theta$$

$$Z_I = Z'' = |Z(\omega)| \sin \theta$$

The impedance plots can be divided to two types, the Nyquist Plot and the Bode Plot. The name commonly used as Cole-Cole plot is for the Nyquist Plot. In the Cole-Cole plot, the imaginary impedance component,  $Z''$ , is plotted against the real component,  $Z'$ , at each excitation frequency. The shape of these plots would be a perfect semicircle or a “flattened” semicircle. The Bode plot is plotted to investigate the absolute impedance,  $|Z|$ , and the phase shift,  $\theta$ , each as a function of frequency. Once the bulk impedance,  $R_b$ , is determined, the value is then used to calculate ionic conductivity of the material.

In this study, ionic conductivity for both the PVA based and PEMA based polymer electrolyte at room temperature was determined using the HIOKI 3532-50 LCR HiTESTER impedance analyzer over the frequency range between 50 Hz and 5 MHz. Ionic conductivity of polymer electrolytes was determined using equation 3.1 below:

$$\sigma = \frac{l}{R_b A} \quad (\text{Equation 3.1})$$

where,

$\sigma$  = electrical conductivity in ( $\text{Scm}^{-1}$ ),

$l$  =the thickness of the polymer electrolytes (cm)

$A$  =the area of the sample in  $\text{cm}^2$

$R_b$  =bulk resistance of polymer electrolytes (determined from the EIS Nyquist plot)

To ensure reliability and reproducibility of the investigation carried out, the ionic conductivity readings for each polymer electrolyte film were repeated three (3) times. The average was then calculated and used to plot the graph. The graph was also plotted with the error bars included.

### 3.4.2 Temperature Dependent Ionic Conductivity Studies

Impedance spectroscopy is also carried out at different temperature to determine the conductivity mechanisms. The experiment would yield conductivity values at different temperatures, which are then plotted in the form of log ionic conductivity against  $10^3/T$ . This graph would help in the conductivity mechanism investigation. If the regression value of all the points in the plot is almost unity, a straight line represents the plot and the conductivity is said to obey the Arrhenius equation as given below.

$$\sigma = (\sigma_0 / T) \exp(-E_a / kT) \quad (\text{Equation 3.2})$$

The mechanism used to describe this mechanism is a thermally activated hopping mechanism. The understanding is that, the vibration of the salt that has complexed with the atom increases with the temperature. This vibration causes the salt to 'hold on' or 'cling' to another free atom when the atoms are close together. If the plot is a curve, then the conductivity can be described by the VTF equation:

$$\sigma = A \exp \left\{ -B / k (T - T_0) \right\} \quad (\text{Equation 3.3})$$

For PVA and PEMA polymer electrolytes, temperature dependent conductivity was also carried out over the frequency range between 50 Hz and 5 MHz from ambient temperature to 120 °C at a signal level of 10 mV. To begin with, a digital micrometer

screw gauge was used to measure the thickness of the polymer electrolyte films. The polymer electrolytes were then sandwiched on a sample holder under spring pressure in the configuration stainless steel (SS) blocking electrode/polymer electrolyte/ blocking electrode to carry out the respective measurements.

As with the room temperature ionic conductivity investigation, to ensure reliability and reproducibility of the investigation carried out, the ionic conductivity readings for each polymer electrolyte film were repeated three (3) times. The average was then calculated and used to plot the graph. The graph was also plotted with the error bars included.

### **3.4.3 Differential Scanning Calorimetry (DSC)**

Differential scanning calorimetry (DSC) is used to study the thermal properties of the prepared polymer electrolytes. Scientists consider this as a very useful instrument from a chemical thermodynamic point as all chemical and physical changes involve enthalpy change and enthalpy is a state function. A DSC instrument can measure the change in heat capacity,  $C_p$ , as a function of temperature. It monitors heat effects associated with phase transition and chemical reaction with relation to temperature. When conducting DSC, the difference in heat flowing to the sample and a reference at the same temperature, is documented a function of temperature. The reference most commonly used is just an empty aluminum pan. To obtain a reading, the temperature for both the reference and the sample would be increased at a constant rate.

In this study, thermal properties of the polymer electrolyte films was determined using TA Instrument Universal Analyzer 200 comprising a DSC Standard Cell FC as main unit and Universal V4.7A software. The procedure was performed in a nitrogen

atmosphere at a flow rate of 60 mL min<sup>-1</sup>. Samples were first weighed to be between 3–5 mg and sealed in aluminum pans. They were then heated from 25 °C to 105 °C at a heating rate of 10 °C min<sup>-1</sup> to remove trace amount of moisture. The heating process was maintained at 105 °C for 5 minutes to ensure complete evaporation. The samples were then cooled to 25 °C, achieving equilibrium and then reheated to 110 °C followed by with rapid cooling to –50 °C at a heating rate of 10 °C min<sup>-1</sup>. The final heating scan was used to evaluate the glass transition temperature ( $T_g$ ).

#### **3.4.4 Linear Sweep Voltammetry (LSV)**

Linear sweep voltammetry is a technique where the cell current at a working electrode is measured as function of time and as a function of potential between the working electrode and reference electrode. A fixed potential range is used and the scanning is done from a lower limit to an upper limit.

Ionic liquid–free polymer electrolyte and the most conducting ionic liquid added polymer electrolyte was subjected to LSV to study the electrochemical potential window using CHI600D electrochemical analyzer. The voltage range studied was ±3V. The polymer electrolyte was placed between SS electrodes at a scan rate of 10 mVs<sup>-1</sup>.

#### **3.4.5 X-Ray Diffraction (XRD)**

XRD can be used to prove that complexation takes place between host polymer, the salt and additives. Prior to obtaining the x-ray diffractogram of the complexed polymer electrolyte, we first obtain the x-ray diffractogram of the pure polymer, pure salt and pure ionic liquid. The shift of the peaks in the pure host polymer, the salt and the ionic liquid is compared to the complexed polymer-salt-ionic liquid and the existence of new peaks would prove the occurrence of complexation.

X-ray diffraction occurs when the monochromatic beam of X-ray interacts with a sample, which then causes the X-rays to scatter from the atoms of the target sample. When the structure is crystalline, the X-rays can undergo constructive or destructive interference and this is what is called diffraction. This process can be described by Bragg's Law,

$$n\lambda=2d \sin \theta \quad \text{(Equation 3.4)}$$

Bragg's Law relates the wavenumber of the electromagnetic radiation to the diffraction angle and the lattice spacing in the crystalline sample. All samples are scanned at a range of  $2\theta$  angles to ensure all possible diffraction directions of the lattice could be achieved due to the random orientation of the powdered material.

XRD technique can also be used to investigate whether a material is amorphous or crystalline. If it is crystalline, the addition of a salt or ionic liquid may make the final material more amorphous. A more amorphous material would produce a higher ionic conductivity due to the flexibility of its backbones, which allow greater polymer flow and ionic diffusivity.

In this research, XRD is used to determine the crystalline/amorphous nature of the polymer electrolytes. The analysis was performed with a BTX benchtop X-ray diffractor using Cu K $\alpha$  radiation wavenumber  $\lambda = 1.5418 \text{ \AA}$  for  $2\theta$  angles between  $5^\circ$  to  $50^\circ$ .

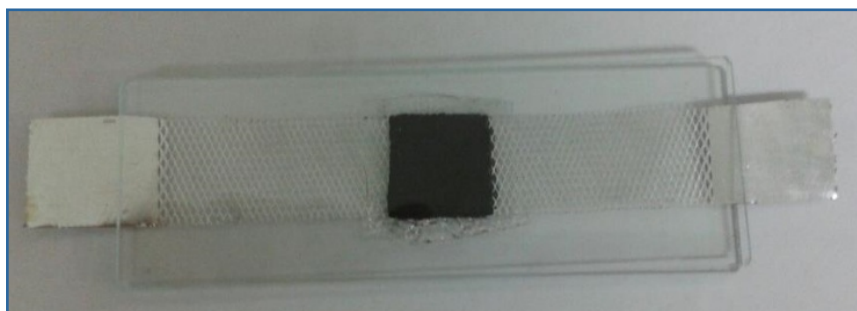


### 3.6 Electrode Preparation

Electrodes for EDLC fabrication were prepared by mixing 80 wt.% activated carbon (Kuraray Chemical Co Ltd., Japan), 5 wt.% carbon black (Super P), 5 wt% multi-walled carbon nanotubes (CNTs) (Aldrich, USA) and 10 wt.% poly(vinylidene fluoride) (PVdF) binder (molecular weight of 534000  $\text{gmol}^{-1}$  from Aldrich) and dissolving them in 1-methyl-2-pyrrolidone (Merck, Germany). The mixture was stirred to obtain homogenous slurry. The slurry was then coated onto the aluminum mesh and oven dried at 110°C. Subsequently, the electrolyte, in its liquid form, was spin coated on the surface of electrodes with a spinning time of 60 secs at 2000 rpm. This was repeated 10 times to ensure the carbon pores are filled with electrolytes as well as to enhance interfacial contact between electrolytes and electrodes.

### 3.7 EDLC Fabrication and Characterization

EDLC cell was constructed in the configuration of electrode/polymer electrolyte/electrode. The prepared EDLC is as in Figure 3.2.



**Figure 3.2: The fabricated EDLC using the highest conducting ionic liquid-based polymer electrolyte from each system.**

The EDLC cell configuration was eventually placed in a cell kit to investigate the electrochemical stability and charge-discharge cycling. Cyclic voltammetry (CV) was carried out to determine the electrochemical stability of the EDLC. The EDLC cell was evaluated at 10 mVs<sup>-1</sup> scan rate in the potential range between 0 and 1 V in intervals of 0.001 V and the specific capacitance ( $C_{sp}$ ) evaluated using this equation [16, 17]:

$$C_{sp} = \frac{i}{sm} (\text{Fg}^{-1}) \quad (\text{Equation 3.5})$$

where,

$i$  = average anodic–cathodic current,

$s$  = potential scan rate,

$m$  = average mass of electrodes which is around 0.01 g.

Galvanostatic charge-discharge was carried out using a Neware battery cycler. EDLC was charged and discharged at a current of 1 mA. Specific discharge capacitance ( $C_{sp}$ ) was obtained from charge–discharge curves, depicted in the equation below [17]:

$$C_{sp} = \frac{I}{m \left( \frac{dV}{dt} \right)} \quad (\text{Equation 3.6})$$

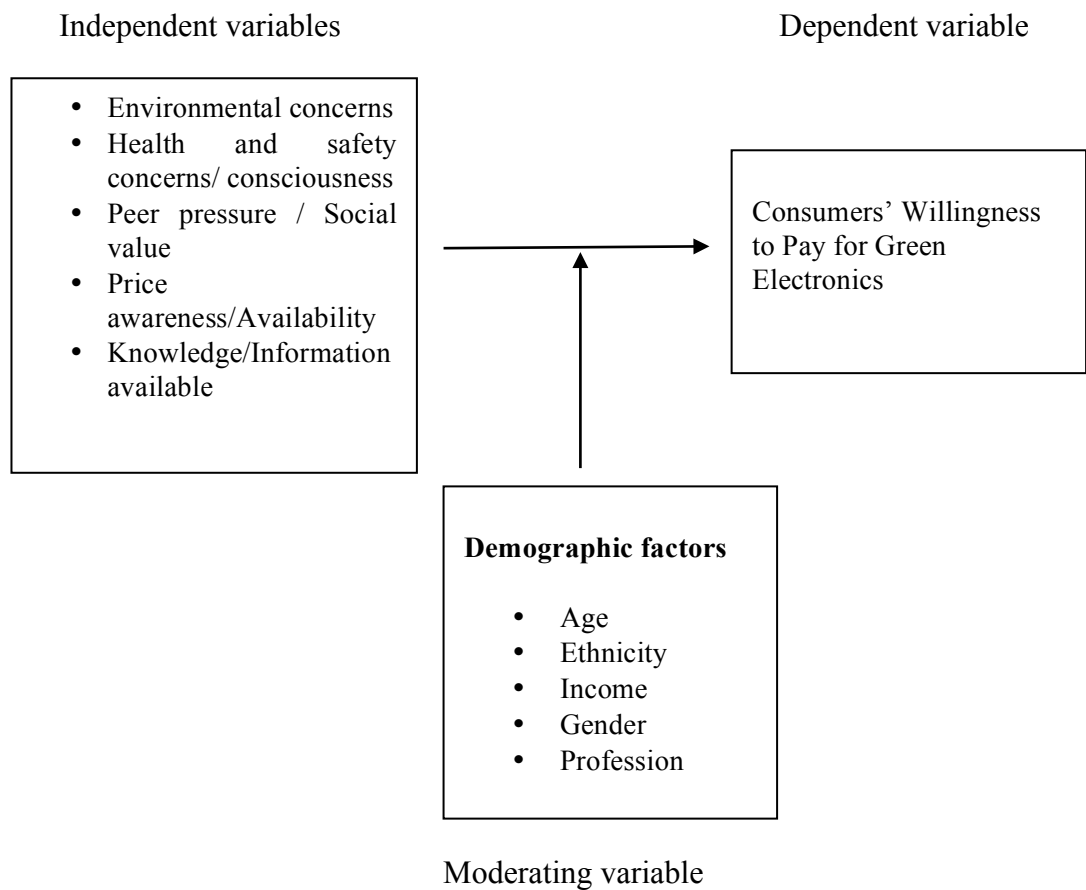
where  $I$  is the current,  $m$  is the average mass of electrode materials,  $dV$  represents the potential change of a discharging process excluding the internal resistance drop occurring at the beginning of the cell discharge and  $dt$  is the time interval of discharging process. The  $\frac{dV}{dt}$  is determined from the slope of the discharge curve.

### **3.8 Survey Data**

The purpose of the second part of the study is to determine the factors that influence consumers' willingness to pay for green electronics. The determinant factors used were obtained through excessive reading and review of literature. Consumers were given all necessary information about green cellphone and the hazards in non-green cellphones. They were then asked how much they would be willing to pay for a green cellphone after asking a series of questions about their environmental and personal knowledge.

#### **3.8.1 Theoretical Framework for the Survey**

Figure 3.3 shows the theoretical framework that we used for the study. The dependent variable in the study is Willingness to pay for green electronics. This study has 6 independent variables, which were decided upon after in-depth literature review and analysis. The moderating variables were an important factor in knowing the socio-demographic background our respondents came from.



**Figure 3.3: Theoretical Framework of Study**

### **3.9 Research Design**

The research was conducted using both secondary and primary data. The secondary data research was conducted to obtain a deeper understanding of the variables, which may be relevant to the study. The primary data is the survey that we conducted to obtain raw data, which we then analysed. As dependent as secondary data may be, it does not always answer the question that is addressed by a particular researcher. It may not be specific to a researchers objective. Due to that, the researchers would need to develop their own method to obtain data to fulfil the purpose of the research and to answer the research questions and achieve its objectives.

Two methods can be incorporated to obtain data – the qualitative and quantitative method. Qualitative research method involves collecting information, analysing and understanding the data by observing respondents reactions (Babin & Zikmund, 2015). It closely observes the human nature and distinctive reactions. The data is unstructured and there are no formulas or numericals but more of observing, interviewing and analysing. Quantitative research, on the other hand, involves empirical evaluation of numerical measurement and analysis. The most widely used method of quantitative analysis is the survey method in which a large number of respondents would participate. The study is usually structured and the survey would include a set of pre-determined research questions, which need to be answered by the survey respondents. The analysis of the respondents' replies would lead up to answering the objectives of the study. Quantitative research would also involve a theoretical or a conceptual framework that includes the types of variables used for the study and the relationship between the variables to the analysed subject. Upon obtaining the survey data, the results are tabulated and analysed using statistical methods which would then be used to decide if a variable is significant in the study or not.

### **3.10 Willingness to Pay Measurement Method**

Willingness to pay is an economic term and the measurement of willingness to pay has been discussed in the economic world from time to time. Several methods have been suggested and improvements are still being made on the methods. The most appropriate method to be used for measurement would depend on the type of data that is obtained from a survey. Whether the data is from actual choices of existing goods or hypothetical items. The most used methods to obtain consumers' willingness to pay are contingent valuation, experimental auctions and choice experiments. In our study we will be using the contingent valuation method.

### **3.10.1 Contingent Valuation (CV)**

Contingent Valuation is a method that is used to determine the monetary value of non-market goods and services. This means that the product has not yet been commercialized. As there is no market price for the goods we are proposing, this method creates a hypothetical market and asks respondents how much they would be willing to pay for goods or services by using a survey questionnaire. This method can also be equated to a market survey that is done before a product comes out for sale to investigate consumers' response and demand for the particular product, cost wise. Most of the time, this method is used to elicit willingness to pay for environmentally related products or services. This method has also been used to investigate willingness to pay for organic food (Sriwaranun et al., 2015).

Our study uses CV to elicit consumers' willingness to pay for green cellphones in Malaysia. Green cellphones are not yet in the market but research and development is rampantly being undertaken to ensure the product that comes to the market one day is equal in efficiency with the existing conventional cellphones. This would be the ultimate goal of telecommunications companies: to obtain an affordable green cellphone to promote environmental sustainability.

The format that we use to determine how much consumers' were willing to pay 'ONCE' for a green cellphone was an amount range where consumers' pick the range they would be willing to pay.

### **3.11 Research Instrument**

As mentioned earlier, the survey questionnaire was developed upon study relevant literature on the green electronics and consumers' willingness to pay for green

products (Ha & Janda, 2012; Mohd Suki, 2016; Sriwaranun et al., 2015; Suki, 2013). This questionnaire collected information on Malaysian consumers' environmental awareness and concerns, their attitudes towards the environment and health and their willingness to pay for green cellphones. The major determinants used for willingness to pay was environmental concern, price awareness, available information on the product, health and safety concerns, peer pressure, availability of the product and their socio-demographic information. The questionnaire deployed mainly open-ended questions and several close-ended questions and the scale used was the 5 – point Likert scale.

There were three parts to the questionnaire. Before the survey proper, consumers' were first informed of the hazards of non-green cellphones and the toxic elements it contains. The first part was to reveal the intensity about consumers' behaviour and attitudes pertaining to the environment, health and safety and how information or availability of a product influences them to purchase a product. There were six sub- sections in the first part. The first sub-section asked about consumers' environmental awareness and if they thought that the current environmental conditions were a reason for concern. The second sub-section was on health and safety concerns to find out how the consumers' valued their health and their thoughts on environmentally safe products with relation to health. The third sub-section was to determine if the consumers' friend or peers in any way influenced the green consumption of the consumers'. The fourth and sixth sub-sections were related because it asked about whether information and availability of a particular product is important for consumers' to decide whether or not they are going to purchase a green product. The fifth question was on asking consumers' whether or not price would affect their decision to purchase a green product or not.

The second part of the survey was the willingness to pay part. This is the CV part of the study. In this part the consumers' were first asked whether or not they would purchase a green cellphone. If the consumer said 'Yes', a follow up question was asked asking the consumers' to indicate the amount they were willing to pay. For those that said 'NO', three options were given to answer why they would not want to purchase one.

The third part of the study was socio-demographic profile of the consumers. One of the questions asked was the household income. This was important, as the household income is believed to reflect the consumers' ability to pay.

The questionnaire uses a Likert scale to measure the responses in the first part of the study. This scale is commonly used in social science study and can range from a 4-point scale to a 7-point scale. This study uses a 5-point scale from 1-5 where 1= strongly disagree, 2= disagree, 3= undecided/neutral, 4= agree and 5= strongly agree to measure the constructs.

This study was a self-administered study, which was developed using Google Drive and sent to individuals or groups through emails and other available technology. No paper survey was sent out. The advantage of such Internet surveys is that it is fast and convenient, as the respondent will have the flexibility to complete the questionnaire at his or her convenience and definitely cost effective. In addition, the respondent and researcher would not have any communication, which removes bias. With the help of Google Drive the results are collected and tabulated as it comes in and that researchers would be able to monitor the number of respondents who have responded to the survey. At the same time, an Internet survey would mean there would be no opportunity for follow-up questions or interviews. The research instrument used is in Appendix 1.



### **3.12 Pre-test and Pilot Study**

A pre-test of the survey questionnaire is important although not compulsory before disseminating the questionnaire. It can help improve the survey results by ensuring that the questions can be understood by the respondents and to ensure the language used is easily understood. This questionnaire did not exactly undergo a pre-test but it was carefully discussed and checked with the supervisor and academicians.

A pilot study was conducted among 20 participants. The objective of a pilot study is to ensure that respondents understand the questions in the questionnaire which would signify the reliability of the data. Of the 20 questionnaires that were given out, all 20 were received with completed replies. This showed that respondents understood clearly what was asked in the questions and there was no need to adjust the questions in the questionnaire.

### **3.13 Sample selection**

It is important for a study to select a sample from a population. This sample would then reflect the target population that the study, which would help the researcher to draw a conclusion about the whole population. As it would be impossible for a researcher to include a whole population in a study, a sample would suffice, as the sample would be selected according to the criteria of the research. Another important point is that the sample size that is selected should be adequate for the survey results to be accurate and reliable for conclusions to be drawn.

The target population of the study was working Malaysians, regardless of gender, ethnicity and geographical location. The method of sampling used was convenience sampling because we already had a target population. The sample size used

was obtained from Table 3.4 (Hair et al., 1995; Krejcie & Morgan, 1970). Based on the table the minimum number of respondents for our study should be approximately 384. In addition, the researcher stated that for larger populations, the sample size required does not increase at a proportionate rate.

**Table 3.4 :Table for Determining Sample Size From A Given Population (Krejcie  
& Morgan, 1970)**

<i>N</i>	<i>S</i>	<i>N</i>	<i>S</i>	<i>N</i>	<i>S</i>
10	10	220	140	1200	291
15	14	230	144	1300	297
20	19	240	148	1400	302
25	24	250	152	1500	306
30	28	260	155	1600	310
35	32	270	159	1700	313
40	36	280	162	1800	317
45	40	290	165	1900	320
50	44	300	169	2000	322
55	48	320	175	2200	327
60	52	340	181	2400	331
65	56	360	186	2600	335
70	59	380	191	2800	338
75	63	400	196	3000	341
80	66	420	201	3500	346
85	70	440	205	4000	351
90	73	460	210	4500	354
95	76	480	214	5000	357
100	80	500	217	6000	361
110	86	550	226	7000	364
120	92	600	234	8000	367
130	97	650	242	9000	368
140	103	700	248	10000	370
150	108	750	254	15000	375
160	113	800	260	20000	377
170	118	850	265	30000	379
180	123	900	269	40000	380
190	127	950	274	50000	381
200	132	1000	278	75000	382
210	136	1100	285	100000	384

Note.—*N* is population size.  
*S* is sample size.

The formula is as follows  $s = \frac{X^2 NP (1-P)}{d^2 (N-1) + X^2 P (1-P)}$ .

*s* = required sample size.

$X^2$  = the table value of chi-square for 1 degree of freedom at the desired confidence level.

*N* = the population size.

*P* = the population proportion.

*d* = the degree of accuracy expressed as a proportion (.05)

### **3.14 Data Analysis**

The data analysis in this study was carried out using the (Statistical Package for Social Sciences) version 12.0 software. For the willingness to pay models, Logistic Regression was used. There are several conditions to be able to use Logistic Regression in a study as will be described below.

Most statistical analysis use linear regression as it is more straightforward. However, there are limitations of linear regressions that make it unable to be used for certain types of data. The major limitation is that linear regression cannot be used to analyse dependent variables, which are dichotomous and categorical. In our study, the dependent variable is willingness to pay and is dichotomous. Consumers' are either willing to pay or not willing to pay, which is categorized by '1' or '0'. Logistic regression is commonly used in studies where there are numerical and categorical independent variables as well.

In a nutshell, it can be said that Logistic Regression determines the impact of several independent variables at one time to predict the response to the dependent variable. Logistic regression utilizes the binomial probability theory to predict the probability, that the probability is either 1 or 0. It uses the maximum likelihood method to form the best fitting equation, which then helps to classify the data we have obtained. Just like normal regression, logistic regression too provides regression coefficients, 'b', which would predict each of the independent variables contribution to the dependent variable.

Logistic regressions is used for two main reasons:

- To predict group membership – as logistic regression tabulates the probability of positive (Yes) over negative (No), the results of the analysis would be presented in the form of Odds Ratio (OR)
- To provide information on the strength of the relationship between the variables.  
For example, in our study, peer pressure increases the probability of willingness to pay for green electronics.

For a study to be eligible to use Logistic regression, there are 4 assumptions that need to be fulfilled.

- The dependent variable need to be measured in a dichotomous scale
- Your study should have more than one independent variable, which is either categorical (nominal or ordinal variable) or continuous (interval or ratio variable). Continuous variable are like exam marks (measured 0-100), intelligence (IQ scores). Examples of ordinal variables are those using scales (e.g. strongly agree, agree, neutral, disagree and strongly disagree) to describe how a consumer agrees to green cellphones. Nominal variables are variable like gender, race, current job and so on. There is no necessity for the independent variable to be normally distributed or linearly related.
- The groups must be mutually exclusive and exhaustive.

- The number of sample need to be larger than required for linear regression as there needs to be a ‘linear relationship between the continuous independent variable and the logit transformation of the dependent variable’

### **3.15 Survey Limitations**

The study may have certain bias issues. One reason is due to the sampling method used, which is convenience sampling. In this method, the online survey form was sent out and shared among the people mainly the government and private sections. Our sample population which consists of many participants from the government sector may affect not portray the Malaysian population. In the study, we are asking about an item that is not yet in the market. Respondents may answer the best answer because they may not be in the situation yet. Hence, their actual behavior may differ from the way they have answered. As this was an online survey, we choose to believe that participants were not in any form of stress in completing the survey as it was a matter of choice and it could be done at the respondents own time and from his or her own device.

### **3.16 Assumed Research Hypothesis**

Before conducting the study, we came up with several assumptions or research hypothesis:

1. Environmental concerns is positively related to willingness to pay for green electronics
2. Health and safety consciousness is positively related to willingness to pay for green electronics
3. Peer pressure / social value is positively related to willingness to pay for green electronics

4. Availability is positively related to willingness to pay for green electronics
5. Knowledge/Information available is positively related to willingness to pay for green electronics
6. Price barrier is positively related to willingness to pay for green electronics

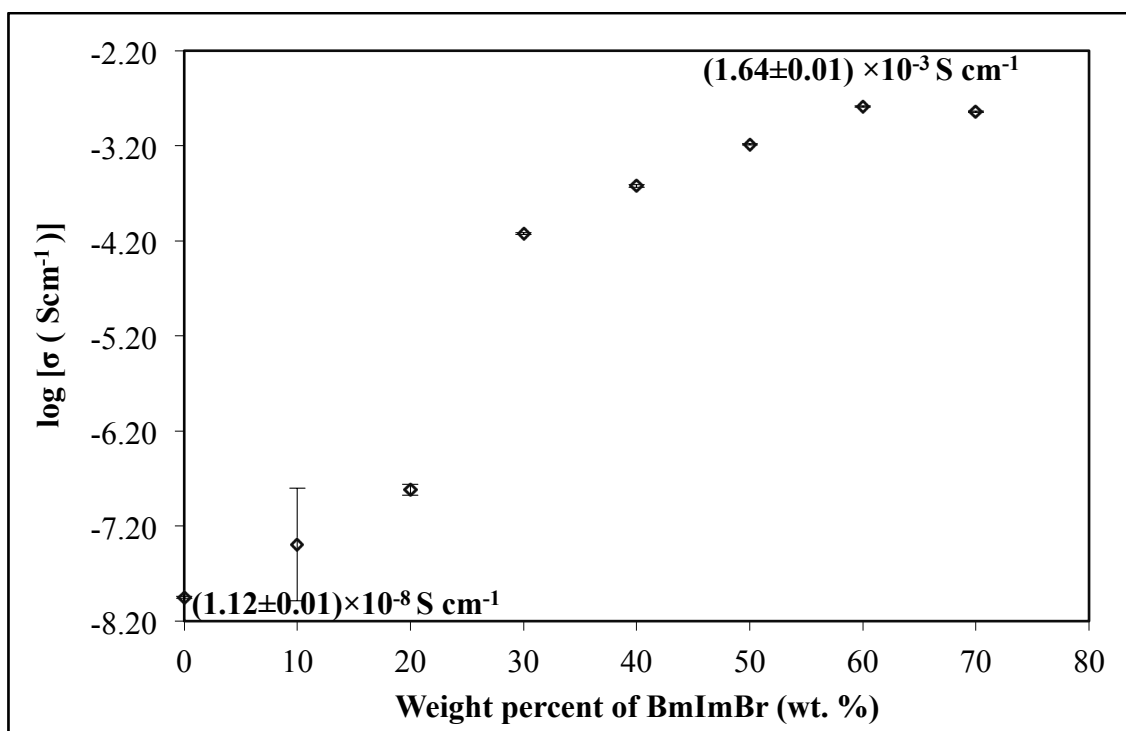
## **CHAPTER 4: RESULTS AND DISCUSSION ON POLY (VINYL ALCOHOL) (PVA) BASED SYSTEM**

This chapter puts forward the results obtained for the poly (vinyl alcohol) PVA, Magnesium triflate salt ( $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ ) and the ionic liquid 1-butyl-3-methylimidazolium bromide (BmImBr) designated as the biodegradable based polymer electrolyte system in our study. This system was designated as biodegradable due to the characteristics of PVA. Poly (vinyl alcohol), unlike most petroleum-based polymers is a proven biodegradable polymer. It has a carbon-carbon single bond backbone, which is completely biodegradable when in the presence of certain microorganisms. In addition, due to the hydroxyl (-OH) functional group is soluble in water and strongly hydrophilic which helps its degradation through hydrolysis (Gohil et al., 2006; Mansur et al., 2008). The polymer electrolyte films were prepared first with salt and then with varied weight % of BmImBr. The films prepared were subjected to ionic conductivity studies to determine the highest ion conducting sample and temperature dependent ionic conductivity studies to study its mechanism of ionic mobility. Then, the polymer films were characterized using Differential Scanning Calorimetry to find out the thermal properties of the films and X-ray diffraction analysis to study the complexation between PVA,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  and BmImBr. Following that, the electrochemical properties of the samples were deduced by Linear Sweep Voltammetry technique. The highest conducting film was then fabricated into EDLC and characterized using Cyclic Voltammetry and Galvanostatic Charge Discharge techniques to examine its performance



#### 4.1 Ambient Temperature Ionic Conductivity Study of Polymer Electrolytes

Figure 4.1 depicts the plot of ionic conductivity of PVA-Mg(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub>-BmImBr polymer electrolyte system with reference to the different weight percentage of 1-butyl-3-methylimidazolium bromide (BmImBr) at ambient temperature up to 70 wt % of BmImBr. Three ionic conductivity data for each sample is averaged to obtain the plotted ionic conductivity value. The error bars are for several plots are small indicating a more certain average value. In addition, the standard deviation obtained for the ionic conductivity values were in the range of 0.01 – 0.09. These low standard deviation values further prove the reliability of the ionic conductivity values obtained.



**Figure 4.1: Ionic conductivity of polymer electrolytes with different weight percentage of BmImBr**

The addition of salt Mg(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub> salt into PVA increases the ionic conductivity value of PVA (pure polymer) from 10<sup>-10</sup> S cm<sup>-1</sup> to 1.12 x 10<sup>-8</sup> S cm<sup>-1</sup>. This is due to the increase in the number of charge carriers in the polymer matrix upon the addition of

magnesium salt (Verma et al., 2014). The increase in the number of charge carriers then increases ion mobility, increasing the ionic conductivity.

Similar results were also obtained by other researchers. The effect of addition of salt on ionic conductivity was also shown by Jeong et al. (2006). In this study of PVA with a variation of salt content with dimethyl sulfoxide (DMSO) as solvent, highest ionic conductivity obtained was  $5.41 \times 10^{-4} \text{ Scm}^{-1}$ . The researchers concluded that the increase in conductivity was not due to free volume behaviour from transport mechanism but due to cations in the polymer electrolytes moving between adjacent ion aggregates which become decoupled from the original polymeric segmental motion. Asmara et al. (2011) in a study with PMMA using  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  in EC and DEC obtained an ionic conductivity value of  $1.88 \times 10^{-3} \text{ Scm}^{-1}$ .

Kumar & Munichandraiah (2002) reported in a study with PMMA doped with  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  and plasticizers EC and PC, that ionic conductivity increases with addition of the magnesium salt to a maximum value of  $4.2 \times 10^{-4} \text{ Scm}^{-1}$ . This increase was attributed to the increase in the concentration of  $\text{Mg}^{2+}$  ion concentration. Other researchers have reported that PEO-magnesium salt complexes result in ionic conductivity ranging from  $10^{-5}$  to  $10^{-4} \text{ Scm}^{-1}$  (Gray, 1991; Levi et al., 2009). It states that the ionic conductivity is restricted due to the restricted mobility of  $\text{Mg}^{2+}$  ions in PEO-Mg salt complexes, as in the PEO- $\text{Mg}^{2+}$  the interaction between the  $\text{Mg}^{2+}$  and the ether oxygen of the PEO is strong thus restricting dissociation of the salt in the polymer matrix (Vincent, 1995).

Ionic liquid was further found to increase the ionic conductivity. Upon addition of 10 wt.% BmImBr, the ionic conductivity shows a gradual increase of about one order

of conductivity compared to ionic conductivity without addition of ionic liquid. The same is observed upon inclusion of 20 wt.% BmImBr. However, upon addition of 30 wt.% ionic liquid, an exponential increase of nearly two orders of ionic conductivity is observed. It can be observed that highest ionic conductivity achieved is  $(1.64 \pm 0.01) \times 10^{-3} \text{ Scm}^{-1}$  at 60 wt.% of BmImBr.

The results obtained are evidence that the number of charge carriers and the mobility of the mobile charge carriers reached its optimum at 60 wt.% BmImBr. Ionic conductivity prior to adulteration and upon adulteration of ionic liquid showed an increase of nearly five orders. This increase in ionic conductivity may be due to the high plasticizing effect of ionic liquids that establishes high ion content (Muchakayala et al., 2018; Tiruye et al., 2016). This plasticizing effect of ionic liquid has the ability to soften the polymer backbone and thus improve the flexibility of polymer chains, which enhances the mobility of polymer segments. This softening of the polymer matrix causes a weakening of the coordinative bonds which then allows the charge carriers to detach from the interactive coordination bonds with high segmental mobility and be mobile, leading to high conductivity of polymer electrolytes (Muchakayala et al., 2018).

It is also due to the plasticizing nature of the ionic liquid that the degree of amorphous entity in the polymer matrix increases. This is explained as the weakening of transient bonds of the molecules in the crystalline region of the polymer matrix, which in turn makes the polymer chains more flexible, thus more amorphous in nature. A more amorphous region means higher mobility of charge carriers in the polymer matrix due to the unordered arrangement of the molecules. In addition, the physicochemistry feature of ionic liquid, namely its low viscosity and high dielectric permittivity also enhances polymer chain flexibility. Furthermore, the thermal stability of an ionic liquid would

depend mainly on the anion, where the cations role in the polymer electrolyte is quite insignificant. Hence, as the anion size increases, the thermal stability too would increase (Leones et al., 2017).

As more ionic liquid is added into the polymer electrolyte, the sticky feature of the polymer electrolyte becomes more apparent. This can be attributed to the plasticizing effect of the ionic liquid. This feature becomes useful during fabrication of an electrochemical device. In the case of this study, electric double layer capacitors (EDLC) were fabricated and for EDLCs its energy storage ability is enhanced by the charge accumulated at both its electrode and electrolyte interfaces. Hence, the better the contact between electrode and electrolyte, the higher the efficiency of the device. The plasticizing effect of ionic liquids will be further discussed in the DSC section.

Upon addition of 70 wt% ionic liquid, the ionic conductivity starts to decrease to  $1.44 \times 10^{-3} \text{ Scm}^{-1}$ . This decrease is highly possible to be due to the agglomeration of ions. When there are excessive ions in the polymer electrolyte, it encourages the formation of ion pairs and ion aggregates which would deter the ionic transportation, consequently decreasing the ionic conductivity. These ion pairs and aggregates would inhibit the conducting pathway and make the backbone less flexible which hampers the ion movement. This would then result in lower ionic conductivity. At 80 wt.% BmImBr, the polymer electrolyte becomes mechanically unstable, hence the inability to obtain ionic conductivity results.

Kumar et al. (2011) found that PEO-Mg<sup>2+</sup> complex gave an ionic conductivity value of  $4.0 \times 10^{-6} \text{ Scm}^{-1}$  at room temperature. Upon addition of ionic liquid 1-ethyl-3-methylimidazolium trifluoromethanesulfonate (EMITf), a maximum ionic conductivity of  $5.6 \times 10^{-4} \text{ Scm}^{-1}$  is achieved for ~50 wt.% addition of EMITf. Upon further addition

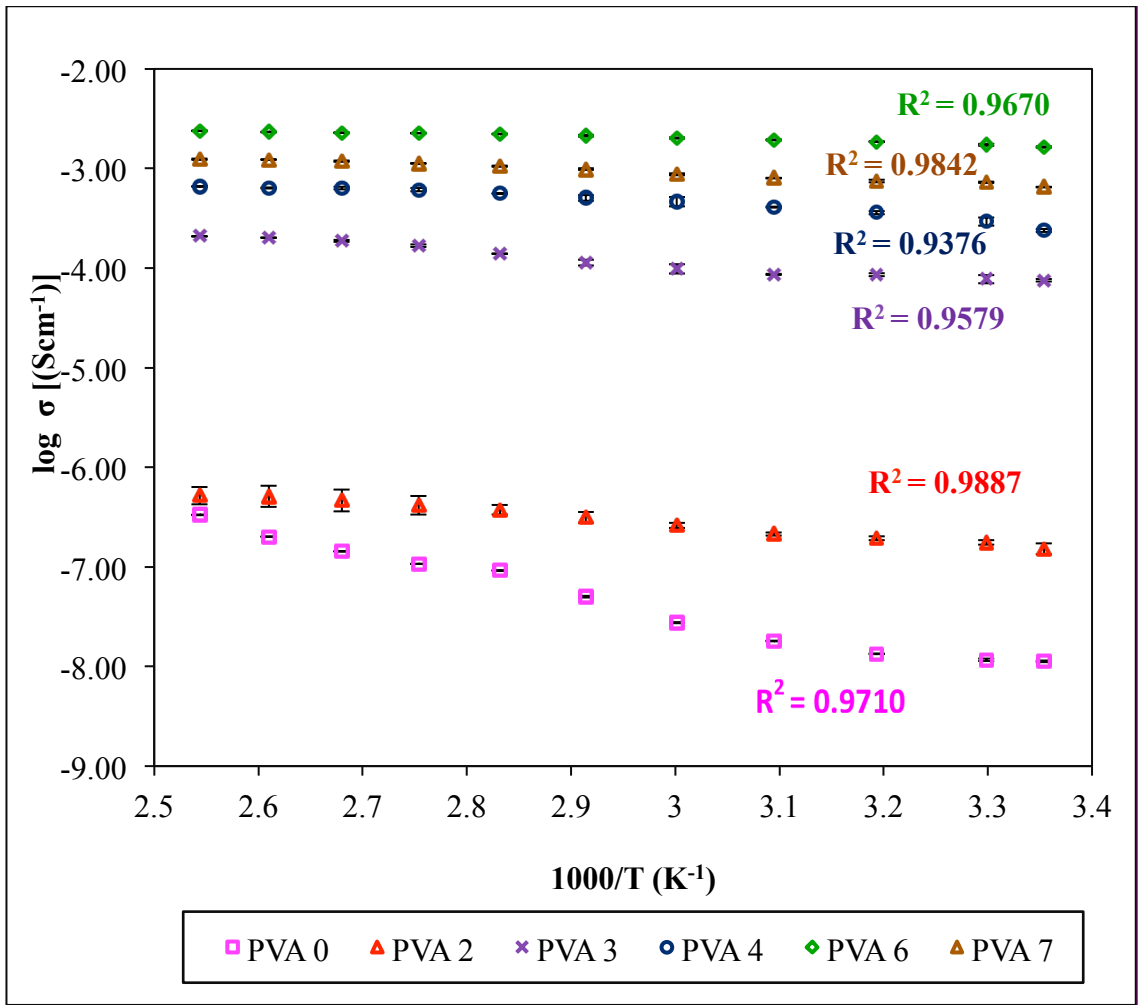
of ionic liquid however, the ionic conductivity was seen to show a decrease. This is the same trend as observed with our study. As expected, the researcher attributed the ionic conductivity enhancement to the effect of plasticization and increase in generation of mobile charge carriers in the polymer matrix which was notably from the ionic liquid with high dielectric constant. The researchers also claimed that in their system with PEO-Mg salts, the contribution by the the Mg salt to the ionic conductivity is close to zero and that it was the addition of ionic liquid that increased the ionic conductivity of the complex. The ionic liquid in their study was portrayed as having two roles, one as an ionic salt and the other as a liquid with low viscosity and high dielectric constant. The first role to assist in enhancing polymer chain flexibility and the second, to dissociate Mg salt and to generate  $Mg^{2+}$  ions to conduct. Hence, we can say that in our study, the addition of ionic liquid did in fact assists in increasing the ionic conductivity of the polymer complex with the BmImBr behaving as a plasticizer and increasing backbone flexibility.

Thanganathan and co-worker prepared PVA based hybrid membranes with phosphomolybdic acid (PMA), 1-butyl-3-methylimidazolium Bis (trifluoromethanesulfonyl) imide (BMITFSI) and 1-ethyl-3-methylimidazoliumtetrafluoroborate (EMI-BF<sub>4</sub>) ionic liquids with SiO<sub>2</sub> inorganic filler. Maximum ionic conductivity of  $0.83 \times 10^{-3} \text{ Scm}^{-1}$  and  $0.58 \times 10^{-3} \text{ Scm}^{-1}$  was obtained for the PVA/PMA/SiO<sub>2</sub>/BMITFSI and PVA/PMA/SiO<sub>2</sub>/EMI-BF<sub>4</sub> hybrid composite membranes, respectively (Thanganathan & Nogami, 2015). The results obtained are also comparable with our results using PVA as host polymer.

## 4.2 Temperature Dependent Ionic Conductivity

Temperature dependent ionic conductivity studies were conducted to determine the mechanism of ionic transport. Temperature dependent plots for ionic conductivity added with different amounts of ionic liquid are fitted with the Arrhenius theory to investigate the ion dynamic mechanism with regards to ions. Figure 4.2 shows the ionic conductivity versus temperature plot of PVA based polymer electrolytes at different weight percentage of BmImBr. The measurement was taken from ambient temperature to 120 °C to ensure we eliminate all water entrapment as distilled water was used as solvent in this system.

Three ionic conductivity data for each sample is averaged to obtain the plotted ionic conductivity value. The error bars for all points are small with some being barely legible at the scale used, indicating a more certain average value. In addition, the standard deviation obtained for the ionic conductivity values were in the range of 0.002 – 0.01. These low standard deviation values further prove the reliability of the ionic conductivity values obtained.



**Figure 4.2: The temperature–dependent plot of polymer electrolytes fitted with Arrhenius equation over wide temperature range**

To identify the mechanism followed by the ions, we first fit the temperature dependence plot with Arrhenius theory as the equation below.

$$\sigma = A \exp\left(\frac{-E_a}{kT}\right) \quad (\text{Equation 4.1})$$

where  $\sigma$  is the ionic conductivity ( $\text{Scm}^{-1}$ ),  $A$  represents a constant which is proportional to amount of charge carriers ( $\text{Scm}^{-1}$ ),  $E_a$  is defined as activation energy (eV),  $k$  is Boltzmann constant ( $8.6173 \times 10^{-5} \text{ eV K}^{-1}$ ) and  $T$  stands for the absolute temperature (K).

The plots demonstrated that the highest ionic conductivity was obtained at 120 °C with 60 wt.% BmImBr added valued  $(2.41 \pm 0.01) \times 10^{-3} \text{ Scm}^{-1}$ . It is noteworthy that ionic conductivity of the polymer electrolyte does show an increase with an increase in temperature, although at a low rate. As temperature increases, movement of polymer chain segments is thermally enhanced accelerating the mobility of the ions. At high temperatures, the vibrations of the molecules increase and the higher the temperatures go the faster the vibrations until the molecules detaches itself from the polymer matrix which is possibly due to the high segmental mobility of the polymer. This in turn would enhance ionic transport since the number of mobile ions would be higher. A combination of increase in temperature and increase in mobile ions would give rise to an increase in ionic conductivity (Kim & Kim, 1999).

From the plots in Figure 4.2, it is notable that the regression values ( $R^2$ ) of PVA 0, PVA 2 and PVA 7 seem close to unity ( $\sim 1$ ) however it cannot be confirmed that they obey the Arrhenius theory. It is also noted that the regression values of PVA 3, PVA 4 and PVA 6 are deviating from unity. To ascertain this, the plots are fitted with volume activated–Vogel–Tamman–Fulcher (VTF) theory using non–linear least square technique. Figure 4.3 depicts the temperature–dependent plot of PVA 0, PVA 2, PVA 3, PVA 4, PVA 6 and PVA 7 polymer electrolytes fitted with Vogel-Tamman-Fulcher (VTF) equation over wide temperature range (ambient temperature to 120 °C). The tabulation of regression values ( $R^2$ ) results is given in Table 4.1 and the empirical equation used to elucidate the results in Table 4.2 is expressed below:

$$\sigma = A_o T^{-\frac{1}{2}} \exp\left(\frac{-B}{T - T_o}\right) = A_o T^{-\frac{1}{2}} \exp\left(\frac{-E_a/k_B}{T - T_o}\right) \quad (\text{Equation 4.2})$$



where  $A_o$  is the pre-exponential constant proportional to the number of charge carriers,  $B$  is a constant determined from the gradient of the plot ( $K^{-1}$ ),  $E_a$  stands for pseudo-activation energy for conduction (eV),  $k_b$  is Boltzmann constant ( $8.6173 \times 10^{-5}$  eV  $K^{-1}$ ),  $T$  represents the absolute temperature (K) and  $T_o$  is ideal vitreous transition temperature at which the polymer segments start to move (Marcilla et al., 2006).  $T_o$  is suggested as 50 K below the glass transition temperature ( $T_g$ ) which is determined from differential scanning calorimetry (DSC) findings (Jiang et al., 2006; Kim & Kim, 1999). These values are tabulated in Table 4.2.

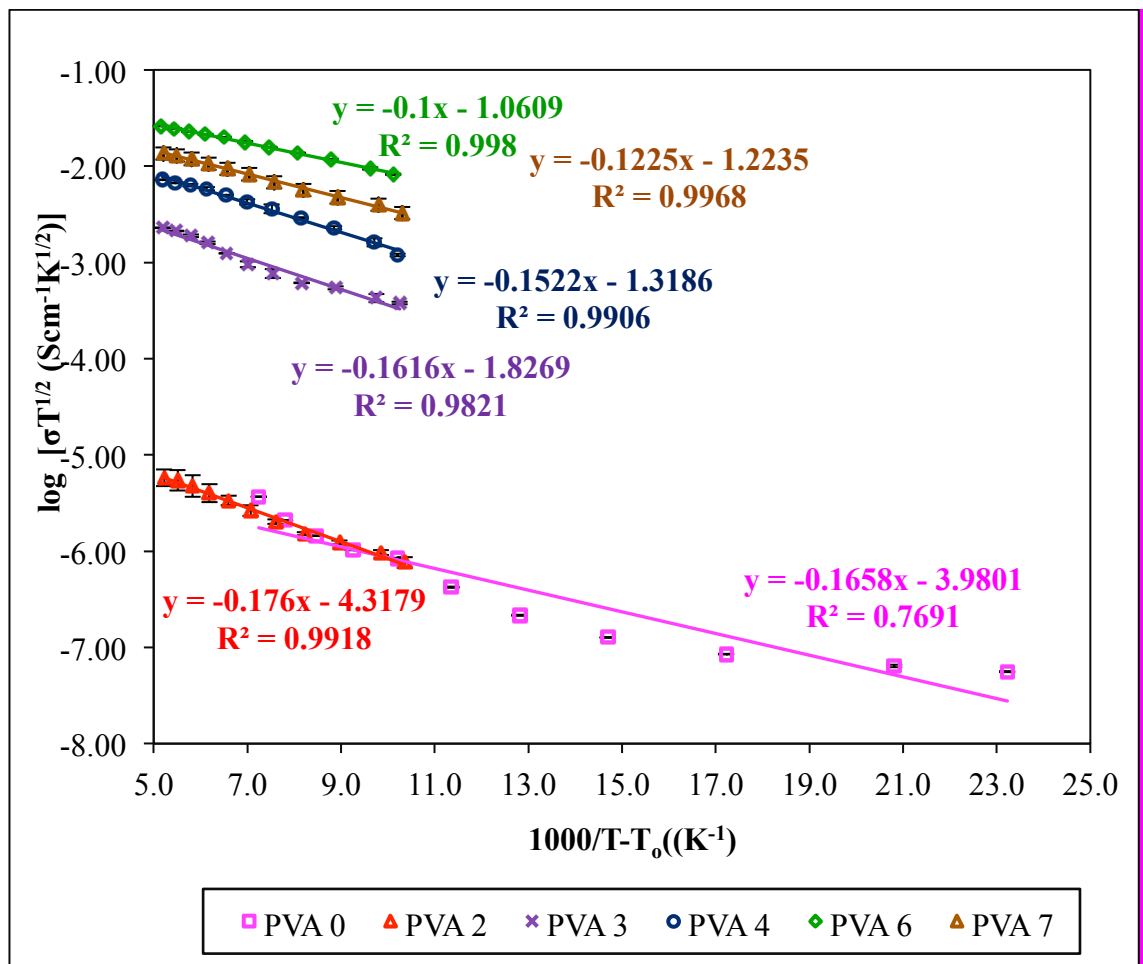


Figure 4.3: The temperature-dependent plot of polymer electrolytes fitted with Vogel-Tamman-Fulcher (VTF) equation over wide temperature range

From both the plots it is noted that only PVA-  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  sample (designated as PVA 0) obeys the Arrhenius theory. All the ionic liquid-added polymer electrolytes exhibit VTF theory associated with the free volume model. The evidence of the VTF rule can be seen from the regression value of all the plots, which approach unity. Hence, we can conclude that these BmImBr incorporated PVA- $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ -BmImBr based polymer electrolytes follow VTF behaviour.

VTF relationship reveals the coupling effect of ionic hopping process with high segmental mobility in an amorphous phase based on free volume theory. At high temperatures, flexibility of the polymer chains is enhanced due to high bond rotation. This high bond rotation coupled with the flexibility of polymer chains would promote the mobility of the ions and generate ionic transportation. High mobility of polymer segments also increases the flexibility of polymer chains due to high bond rotation. High mobility improves ion transport creating more free volume of conducting pathways. Hence, more vacant sites would be available for the charge carriers to be incorporated into the polymer electrolytes, which produce more ion hopping, subsequently increasing ionic conductivity at higher temperatures. The conduction is finally generated through the process above.

Upon carrying out the calculations using the non-linear least square technique, several significant results have been tabulated as below and discussed.

**Table 4.1: The obtained parameters from each VTF plot with the experimental glass transition temperature from DSC thermogram ( $T_0$ )**

Sample	Regression value, $R^2$	Glass transition temperature, $T_g$ (K)	Ideal glass transition temperature, $T_0$ (K)
PVA 2	0.99	251.64	201.64
PVA 3	0.98	250.62	200.62
PVA 4	0.99	250.19	200.19
PVA 6	0.99	249.29	199.29
PVA 7	0.99	251.13	201.13

**Table 4.2: The obtained parameters from each VTF plot ( $A_0$ ,  $B$  and  $E_a$ )**

Sample	Log $A_0$	Pre-exponential constant, $A_0$ ( $\text{mScm}^{-1} \text{K}^{1/2}$ )	Gradient of the plot, $B$ ( $\text{K}^{-1}$ )	Pseudo-activation energy, $E_a$ (meV)
PVA 2	-4.3179	$4.81 \times 10^{-5}$	0.176	34.93
PVA 3	-1.8269	0.014	0.1616	32.07
PVA 4	-1.3186	0.048	0.1522	30.21
PVA 6	-1.0609	0.087	0.1	19.85
PVA 7	-1.2235	0.060	0.1225	24.31

As mentioned earlier, the sample PVA 6 provides the highest ionic conductivity. Upon examining the parameters, this result is clearly evidenced.  $A_0$  is the pre-exponential constant which is proportional to the number of charge carriers and  $B$  is related to the rate which the viscosity changes with temperature (Pandey et al., 2011). From the  $A_0$  values, it is obvious that the highest number of charge carriers is for PVA

6, which correlates with the sample's highest ionic conductivity. It simply means that PVA 6 has the most charge carriers which means higher ion diffusion compared with the other ionic liquid added polymer electrolyte samples. To further investigate the credibility of the aforementioned deductions, the pseudo-activation energy,  $E_a$  of each sample was calculated. It was found that the highest ion conducting sample, PVA 6 displayed the lowest pseudo-activation energy. This can be explained in terms of the flexibility of the polymer backbone added with the higher segmental mobility, which allows the ions to move or hop easily, hence requiring lower activation energy.

Alternatively, the lowest conducting sample PVA 2, shows the lowest number of charge carriers, which relatively mean lowest ionic conductivity. The pseudo-activation energy of PVA 2 was also the highest in comparison, which correlates with its low ionic conductivity.

Incorporation of BmImBr into PVA based polymer electrolyte obeyed the VTF theory. A plausible reason for this may be that free ion number enhances ion mobility, increasing ionic conductivity. In addition, the size of anions also plays a role.  $\text{Br}^-$  with ion size of 190 pm, which is considered a large anion, has self-dissociating properties. This means that due to its large anionic size, the  $\text{Br}^-$  can be easily dissociated from the transient bonding of the anions itself and allows itself to be transported in the polymer matrix. The large anion size would favour polarization thus increasing ionic conductivity in the electrolyte.

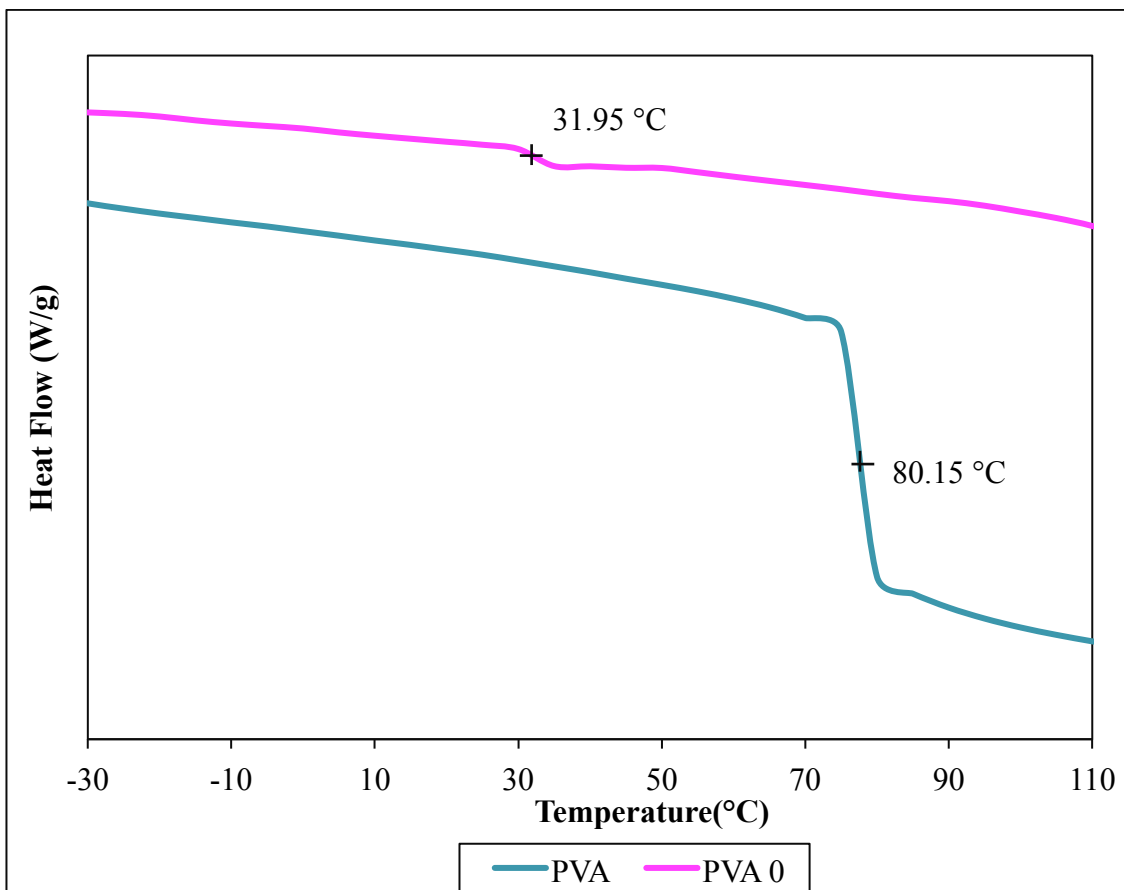
When determining temperature dependent ionic conductivity, it is observed that the highest ionic conductivity obtained was PVA 6 at 120°C at  $2.38 \times 10^{-3} \text{ Scm}^{-1}$ . This can be evidenced due to the addition of ionic liquids and the increase in the ionic

conductivity is ascribed to be because of the plasticizing nature of the ionic liquid. This plasticizing effect of the IL as explained earlier, causes the polymer backbone to soften and become more flexible. This flexibility further enhances the ion movement in the polymer complex.

### 4.3 Differential Scanning Calorimetry (DSC)

Thermal behaviour of the polymer electrolyte was investigated using DSC. For this study, DSC was used to determine the Glass Transition Temperature ( $T_g$ ) of the sample, which is defined as the phase transition of the polymer from the hard, glassy state to an amorphous region, which is a flexible, soft and rubbery state.

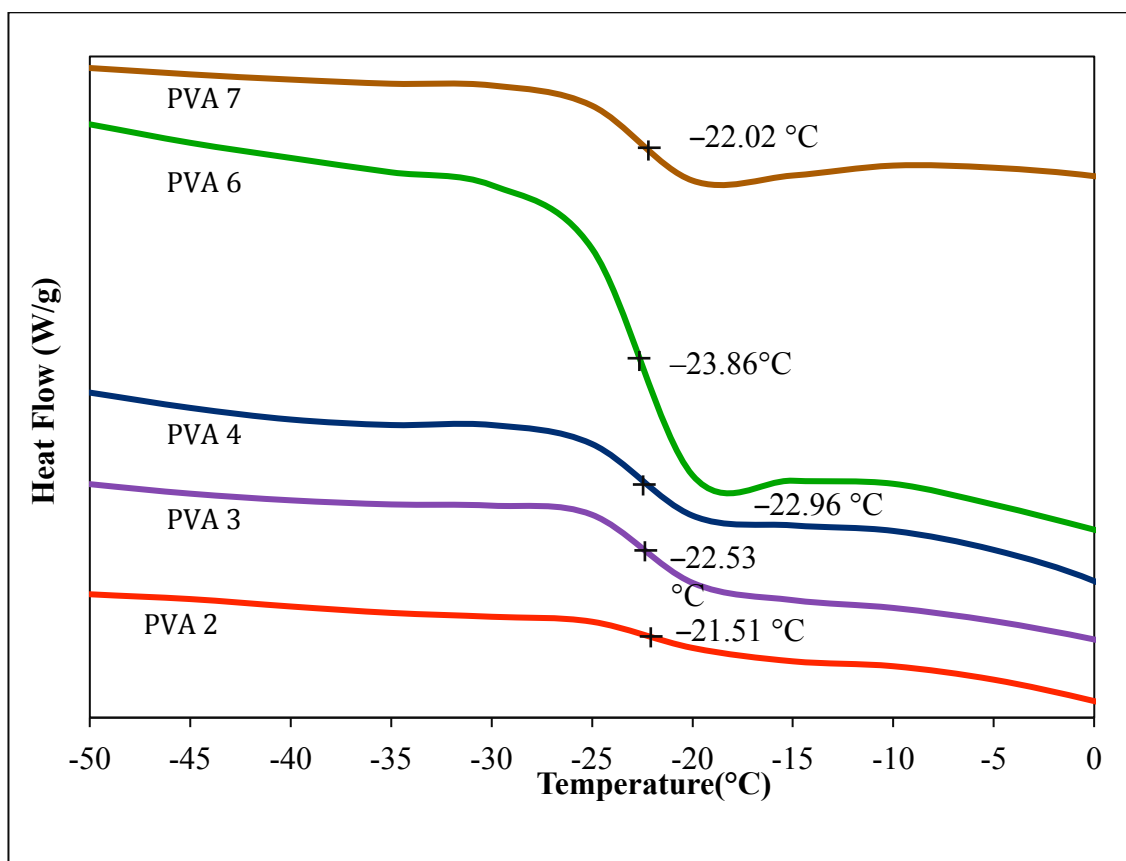
DSC curves of pure PVA and Magnesium Triflate salt added PVA in the temperature range between 30°C to 110°C is depicted in Figure 4.4. The observation is that pure PVA has a  $T_g$  of 80 °C and upon addition of 30 wt.%  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  (PVA 0), the  $T_g$  reduces to 32 °C. This is an endothermic reaction. In addition, there was also no crystalline peak detected in the temperature range studied, substantiating that the reaction took place in an amorphous phase. The amorphous nature of the polymer electrolyte came about due to the  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  used, which provided an add-on plasticizing effect towards the polymer electrolyte, hence exhibiting a lower  $T_g$ . This plasticizing effect thus softens the polymer backbone making it more flexible. This reduction in  $T_g$  also verifies the interaction existing between the salt and polymer. The VTF theory deduces that in the amorphous phase, the ion hopping which is associated with the high segmental mobility of the polymer chains. The amorphous nature is seen through the low  $T_g$  that we obtained experimentally, which explains the rubbery nature of the polymer. Due to that, the polymer backbones are flexible and the charge carriers can move from one site to another empty site.



**Figure 4.4 : DSC thermogram of pure PVA and salt added PVA.**

Figure 4.5 depicts the DSC thermogram for PVA-Mg(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub>-BmImBr polymer electrolytes with different wt% of BmImBr at temperature range between – 50°C to 0°C. It is interesting to note that upon addition of ionic liquid, the  $T_g$  decreases to sub-ambient temperature. The sub-ambient temperature corroborates the transition of the polymer electrolyte to a rubbery phase. At this state, the polymer electrolyte matrix undergoes orientation and conformational changes due to its higher flexibility. From both Figures 4.4 and 4.5 it can be concluded that the backbone flexibility of ionic liquid added polymer electrolyte is much higher than the ionic liquid free polymer electrolyte, which in turn, increases the segmental mobility of the polymer backbone. This is further confirmed through the decrease in  $T_g$  which can be attributed to the plasticizing effect of the ionic liquid. The plasticizing effect of the ionic liquid makes polymer backbone

more flexible by softening the polymer matrix. The ionic liquid would weaken the interaction between the charge carriers coupled with a more flexible polymer backbone favours ion migration throughout the polymer chain. The more flexible polymer backbone enhances faster ion migration and ion conduction giving rise to higher ionic conductivity.



**Figure 4.5 : DSC thermograms of ionic liquid added PVA**

From Figure 4.5, it is observed that the lowest  $T_g$  is exhibited by PVA 6 followed by PVA 4 which corresponds to a  $T_g$  of -23.86 °C and -22.96 °C, respectively. This result substantiates the highest ionic conductivity value which we had obtained for PVA 6. From the lowest  $T_g$  value for PVA 6, we can conclude that the reduction in  $T_g$  means the material is becoming more plastic in nature, which can be attributed to the plasticizing effect of the ionic liquid in the polymer matrix.

Upon addition of 70 wt% BmImBr, the  $T_g$  is found to increase instead of decreasing. This can be attributed to the formation of ion aggregation, which also corresponds to the increasing value of ionic conductivity obtained and discussed earlier. The formation of these ion aggregates would obstruct the flexibility of the polymer backbone, hence impeding the ion migration as well.

#### 4.4 X-Ray diffraction

X-Ray diffraction is a tool to investigate structure of a material that is prepared and identify the phases present. XRD of polymer films was performed to investigate the effect of incorporation of salt and ionic liquid on the crystallinity of the polymer matrix. In addition, XRD will also help us in studying the complexation between PVA,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  and BmImBr. XRD was performed using Siemens D5000 diffractometer with  $\text{Cu-K}\alpha$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ) at 40-kV operation voltage and 30-mA current.

Figure 4.6 displays the XRD pattern of pure PVA,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  salt, BmImBr ionic liquid, PVA-4 and PVA-6. Pure PVA exhibited prominent characteristic peak at  $2\theta$  value of  $19.8^\circ$ , while  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  showed well define crystalline peaks at  $2\theta$  value of  $18^\circ$ ,  $22^\circ$ ,  $23.8^\circ$ ,  $29^\circ$ ,  $30^\circ$  and  $42.9^\circ$  (Chong et al., 2016; Sutka et al., 2015). For BmImBr ionic liquid, very weak and small peaks were observed. It was found that the sharp crystalline peaks of  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  salt disappeared completely when it is added with PVA – ionic liquid complex (evident in PVA-4 and PVA-6). This reveals the successful dissociation and complexation of  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  salt with the amorphous polymer matrix of ionic liquid complexed PVA (Sutka et al., 2015). The amorphous nature is a definite prerequisite for obtaining high ionic conductivity in a polymer electrolyte. In addition, the intensity of the PVA peak was significantly decreased upon incorporation of 40 wt. % and 60 wt. % ionic liquid. The elevated contents of ionic liquid led to increase in the  $\text{BmIm}^+$  ions which effectively weaken the hydrogen



bonding in the PVA polymeric chain resulting in decrease of crystallinity. The increase in amorphous nature can play significant role in conductivity as diffusion of ions can take place very easily. Furthermore, in the amorphous phase, the polymer chains are actually more flexible, allowing an increase in ion mobility, consequently increasing the ionic conductivity of the polymer system. There seems to be conformational changes in the crystalline texture of the PVA due to immobilization of the ionic liquid/Mg-salt in the polymer electrolyte. The smoothing of the peak shows that once the polymer blends with ionic liquid and salt, it acts as a plasticizer to the polymer (Pandey & Hashmi, 2009). The non-existence of additional peaks in the polymer electrolyte matrix imply that the materials used are compatible.

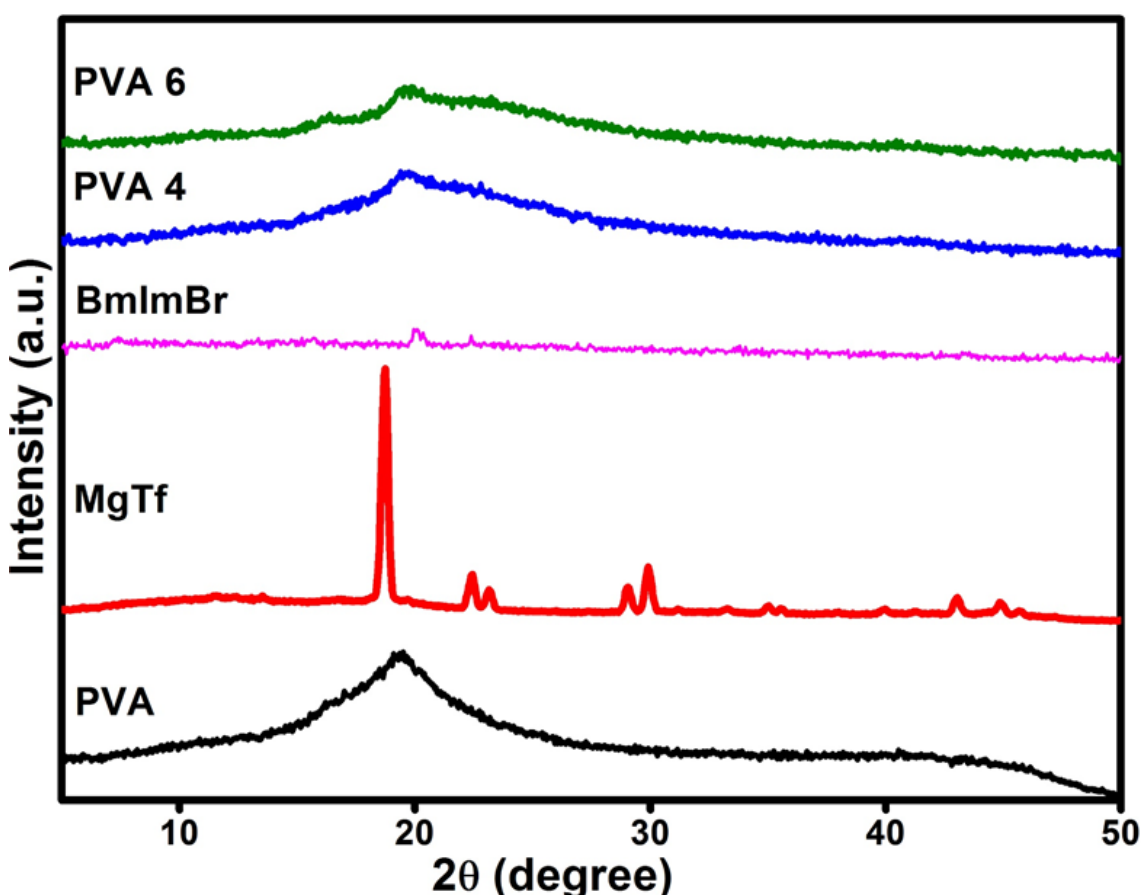
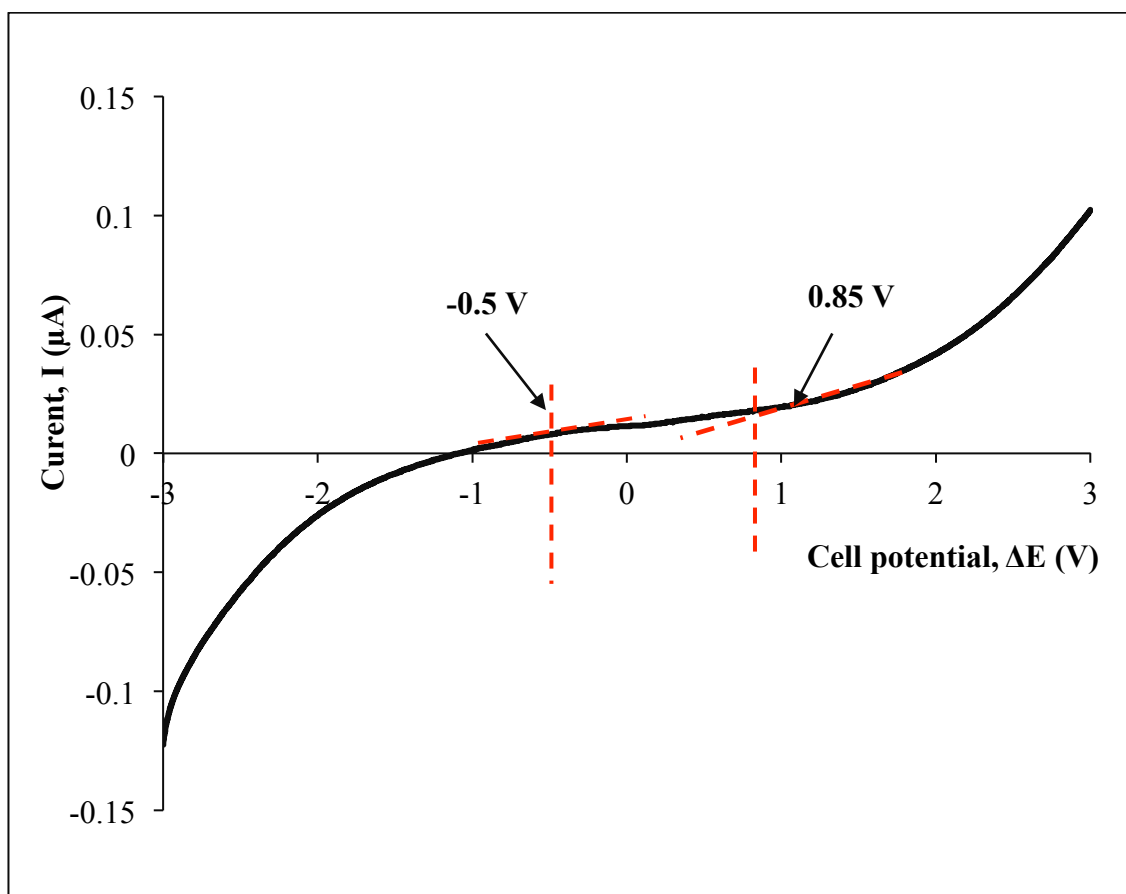


Figure 4.6: XRD patterns of PVA,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  (labelled MgTf), BmImBr, PVA-4 and PVA-6

#### 4.5 Linear Sweep Voltammetry (LSV)

Linear Sweep Voltammetry is carried out to determine the maximum operational potential for a polymer electrolyte for energy storage in an electric double layer capacitor (EDLC). Figures 4.7(a), (b) and (c) represent the potential window of PVA 0, PVA 4 and PVA 6 respectively.



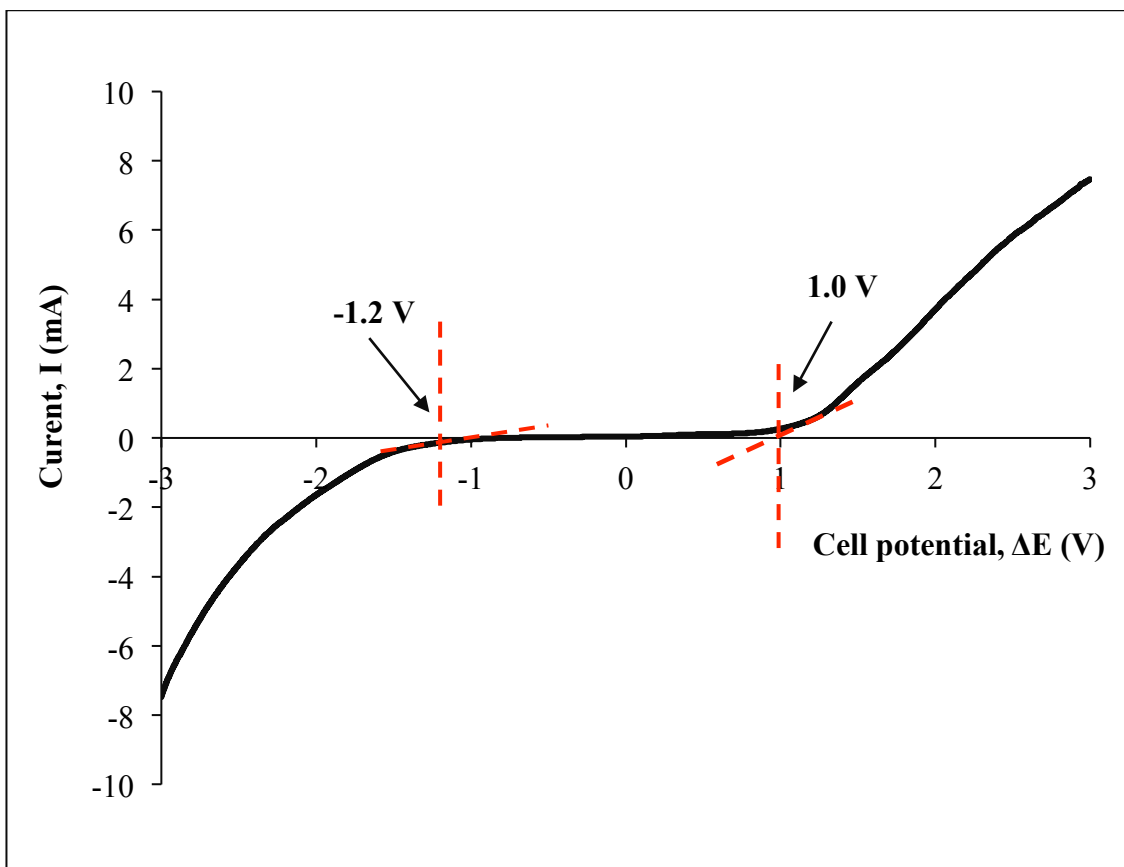
**Figure 4.7(a) : LSV response of PVA 0.**

The LSV of PVA 0 is 1.35 V, where the cathodic and anodic potential are detected at  $-0.5$  V and  $0.85$  V as can be seen from Figure 4.7(a). Upon impregnation with 40 wt% ionic liquid as seen in Figure 4.7(b), the operational potential window widens to 2.2 V between  $-1.2$  V and  $1.0$  V. The reason for this increase is attributed to the higher concentration of ionic liquid in the polymer electrolyte. The LSV for the polymer electrolyte, which showed the highest ionic conductivity was for PVA 6,

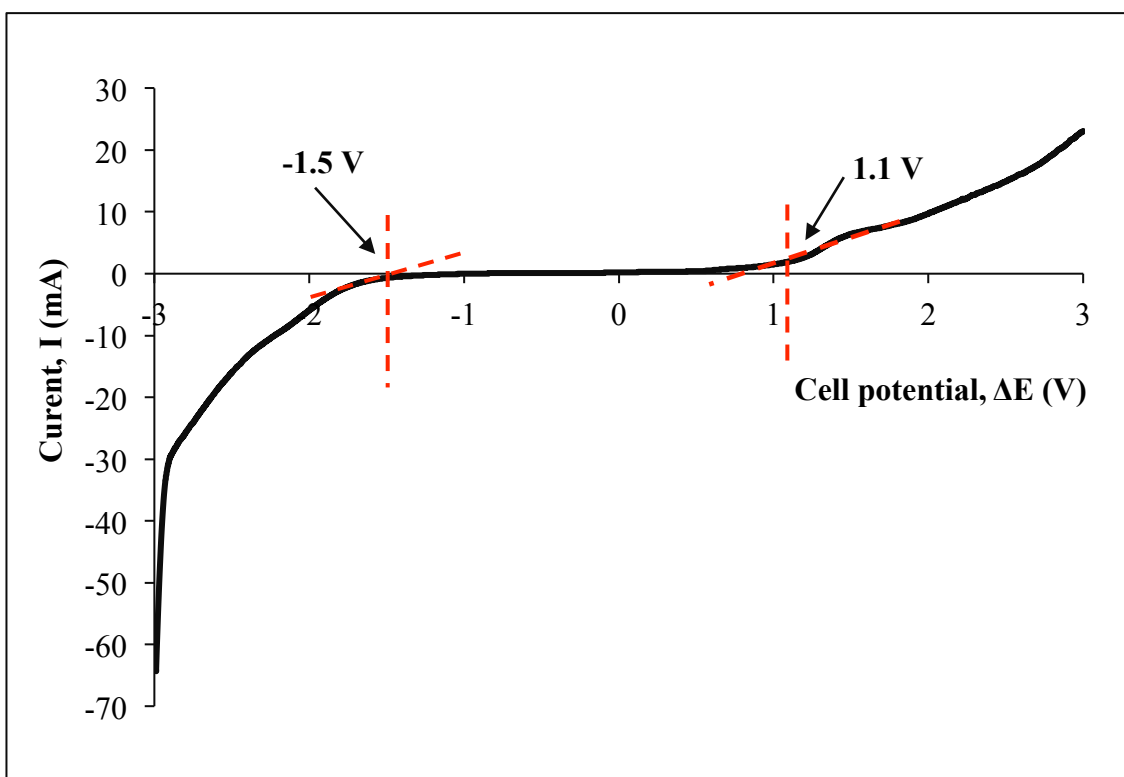
displaying a potential window of 2.6 V. This is clearly depicted in Figure 4.7(c). This further strengthens the postulation that the higher concentration of ionic liquid attributes to this wider electrochemical potential.

As the concentration of ionic liquid is increased, the number of ions increases too. This inspires rapid ion transport. This rapid ion transport mechanism leads to an increase in operational current in PVA 6 compared to PVA 0. The facilitation of ion transportation leads to higher charge accumulation at the electrode – electrolyte interface.

From the tests carried out, it is clear that the potential window for PVA 6 is higher than PVA 4, which is to be expected, as the ionic conductivity of PVA 6 was also the highest. These characterizations pave the way in the decision for selecting PVA 6 as the polymer electrolyte of choice to fabricate EDLC.



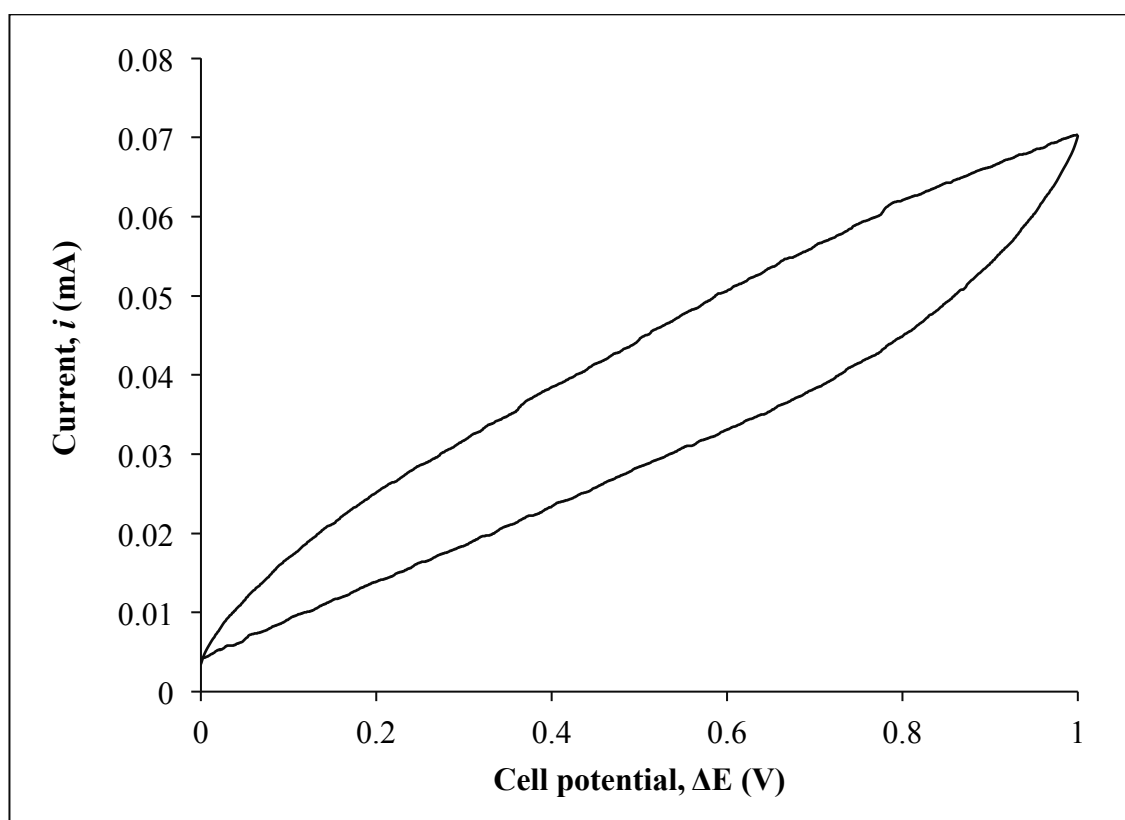
**Figure 4.7(b) : LSV response of PVA 4.**



**Figure 4.7(c) : LSV response of PVA 6.**

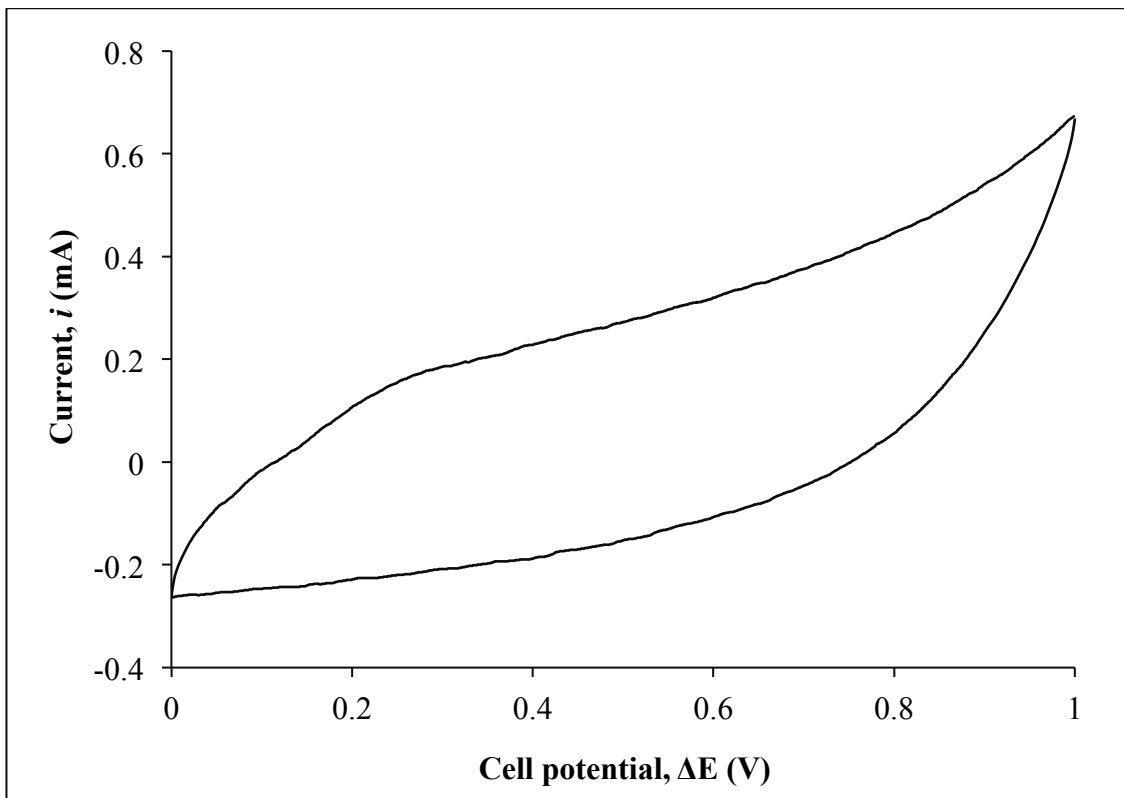
#### 4.6 Cyclic Voltammetry (CV)

Once the polymer electrolyte is used to fabricate the intended device, in this case, EDLC, CV is performed to evaluate the performance of the electrochemical capacitor in relation to its electrochemical behavior and charge storage. As mentioned earlier, the efficiency of the supercapacitor can be investigated through its electrochemical potential window. Figure 4.8 shows the CV curve of the ionic – liquid free EDLC. We can clearly note that the CV results depict the behavior of a Type 1 supercapacitor. This is not an ideal shape for a CV plot of an EDLC. The Type 1 supercapacitor displays a leaf-like shape with a specific capacitance of  $1.41 \text{ Fg}^{-1}$ . This low specific capacitance is due to the poor ionic conductivity of the polymer electrolyte and due to the poor interfacial contact between the electrode-electrolyte.

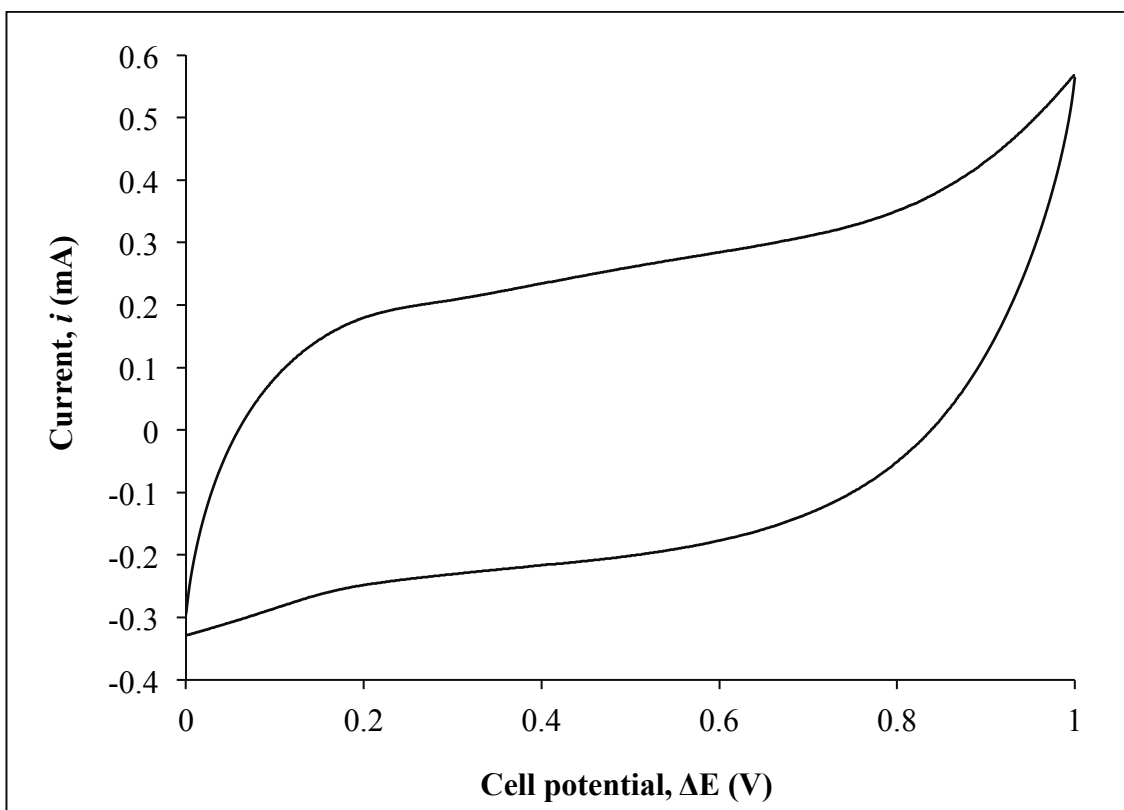


**Figure 4.8 : CV curve of the ionic – liquid free EDLC**

Both Figure 4.9 and 4.10 depict depicts the CV curve of PVA 4 and PVA 6, respectively. PVA 4, the second highest conducting polymer electrolyte shows specific capacitance of  $9.17 \text{ Fg}^{-1}$  and PVA 6, the highest conducting polymer electrolyte, shows specific capacitance of  $45.95 \text{ Fg}^{-1}$ . Comparing the shapes of both the CV voltammogram of PVA 4 and PVA 6, we can see that PVA 6 portrays a rectangular shaped plot. Several reported works are mentioned here. Pandey obtained a specific capacitance of  $2.6\text{--}3 \text{ Fg}^{-1}$  for EDLC using ionic liquid based poly (ethylene oxide) polymer electrolytes and multi-walled carbon nanotubes (Pandey et al., 2010). Shukor et al., (2014) fabricated EDLC using glycerol plasticized-polymer electrolytes and obtained specific capacitance of  $33 \text{ Fg}^{-1}$ .



**Figure 4.9 : CV curve of 40% ionic – liquid added EDLC (PVA4)**



**Figure 4.10 : CV curve of 60% ionic – liquid added EDLC (PVA6)**

A rectangular shape for a CV voltammogram is deemed ideal. The irregular or leaf shape voltammogram in Figure 4.8 shows a Type 1 supercapacitor and its shape can be attributed to the poor contact between electrode and electrolyte which may be due to the absence of ionic liquid. Hence, the poor interfacial contact would hamper the absorption of ions into the electrode surface. In addition, the low ionic conductivity of the ionic liquid free polymer electrolyte also contributes to this effect.

Upon addition of ionic liquid, a notable increase in specific capacitance is observed. The shape of the voltammogram is rectangular, replicating the ideal shape. The addition of ionic liquid gives the polymer electrolyte an adhesive nature, which improves the interfacial contact between electrolytes and electrodes. This leads to better ion absorption and exchange, consequently enhancing the storage ability of the EDLC.

The ionic liquid, which comprises of mainly ions facilitate the ion migration within the polymer matrix. This high mobility and high number of ions cause diffusion of ions into the pores of the porous electrode, accumulating at the electrode-electrolyte boundary which promotes a double layer formation, giving rise to an increase in the energy storage capability of the EDLC.

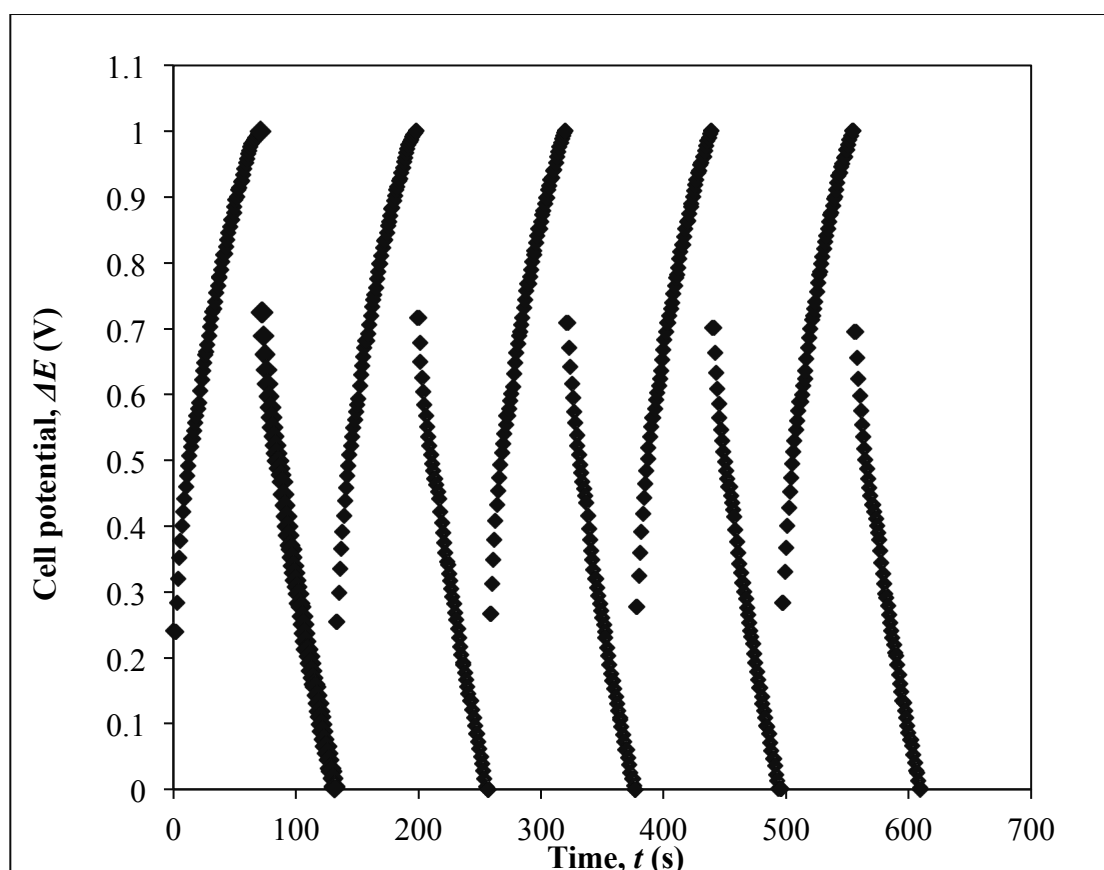
Pandey et al. (2011) showed that using PEO with Li and Mg salts and ionic liquid resulted in low specific capacitance values for both his systems. The Li-based polymer electrolyte offered a capacitance value of  $1.7\text{--}2.1 \text{ F g}^{-1}$ , whereas the Mg-based polymer electrolyte showed improved capacitance of  $2.6\text{--}3.0 \text{ F g}^{-1}$ . However, these values were higher than both systems without ionic liquid which proved the effectiveness of ionic liquid incorporation to improve performance of polymer electrolyte in EDLCs. These authors concluded that the  $\text{Mg}^{2+}$  - ionic liquid PEO-



complex performed better in the fabricated EDLC than the  $\text{Li}^+$  ion system which proves that Mg-based polymer electrolytes are better as a choice for EDLC fabrication.

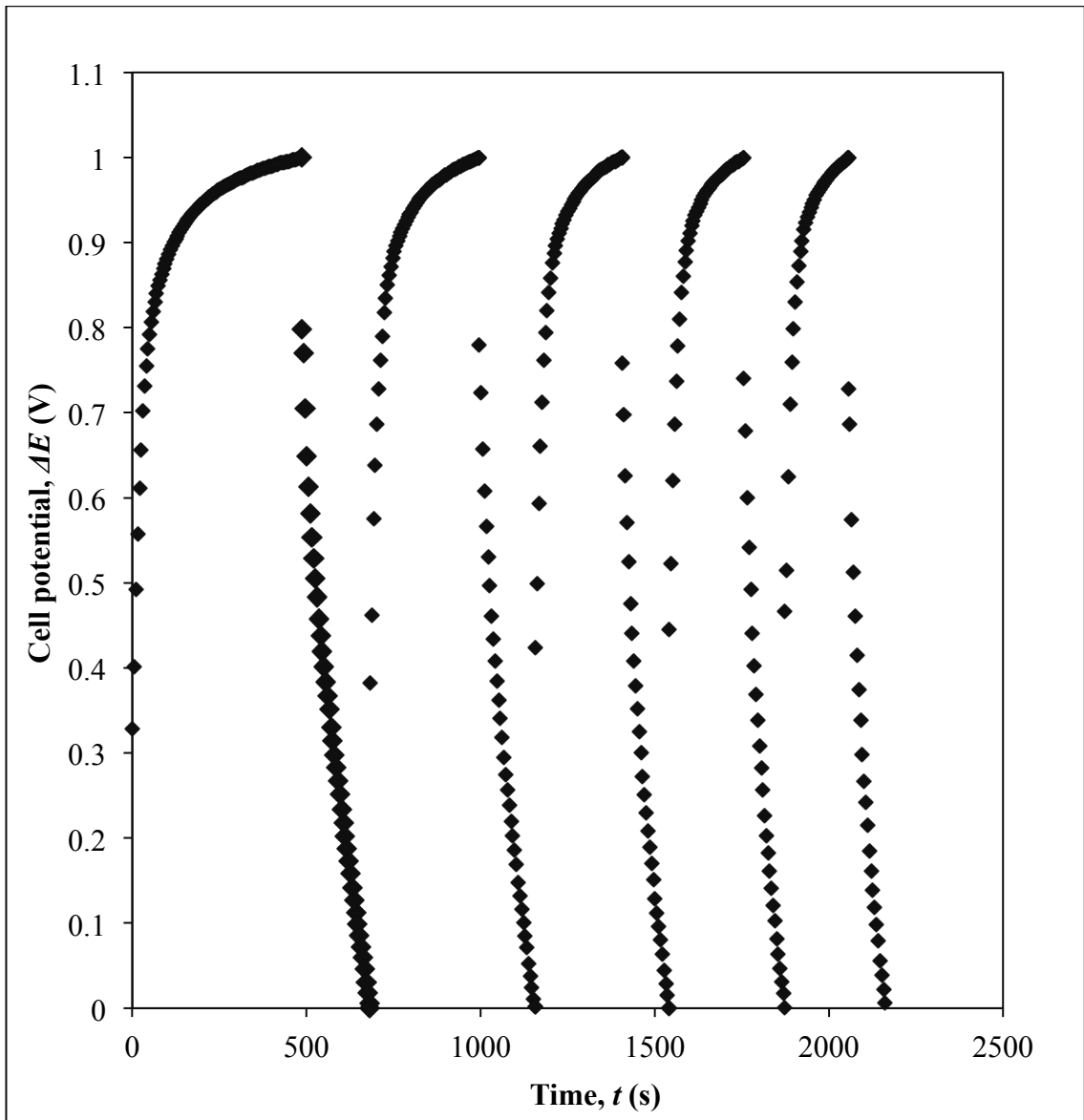
#### 4.7 Galvanostatic Charge Discharge (GCD)

Electrochemical storage and pattern of EDLC can also be evaluated through constant current charge-discharge. The repetitive loop of charging and discharging is known as a cycle. Conventionally, the working voltage of an EDLC is attributed to the electrolyte breakdown voltage whereas electrode resistance and conductivity would affect its internal resistance and capacitance. Figures 4.11 and 4.12 describe GCD for PVA 4 and PVA 6, respectively over 5 cycles of charging and discharging. Additional parameters such as specific discharge capacitance, energy density and power density are also calculated and tabulated.



**Figure 4.11: Galvanostatic charge–discharge performances of EDLC containing PVA 4 over first 5 cycles**

Figure 4.11 depicts a symmetrical shaped charge discharge pattern unlike the Figure 4.12. The symmetrical pattern illustrates the super capacitive nature of the EDLC (Amitha et al., 2009). An ideal case would be for the charging to begin at 0 V to 1 V and to be discharged from 1 V to 0 V. However, it is noted that the charging begins at a slightly deviated voltage. This deviation is attributed to the internal resistance of the EDLC. The figure also depicts a drop at the initial part of the discharge curve, which may be due to ohmic loss, which arises from the resistance between the electrode and electrolyte (Marcilla et al., 2006; Mitra et al., 2001). Ohmic loss can be defined as charge transfer resistance and bulk resistance of polymer electrolyte. Another explanation for ohmic loss may also be attributed to the depletion of polymer electrolyte, which can in turn increase the electrochemical cell resistance.

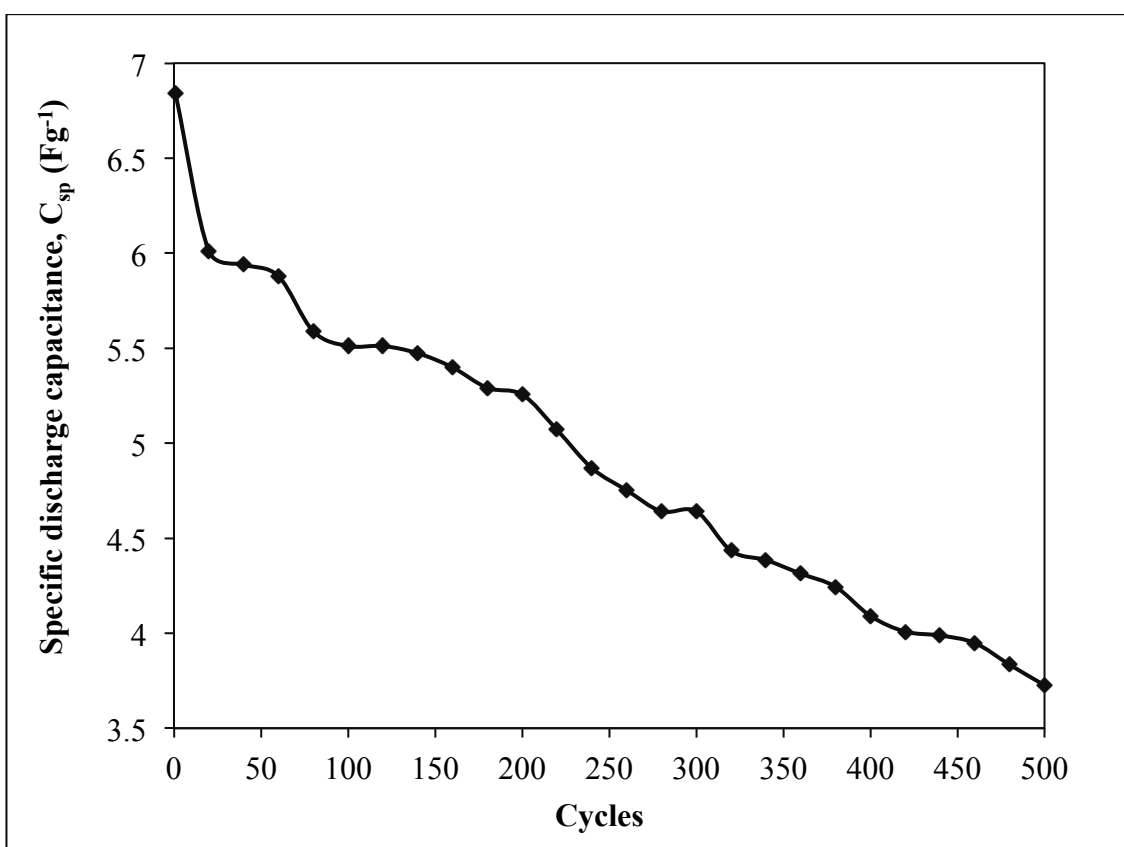


**Figure 4.12: Galvanostatic charge–discharge performances of EDLC containing PVA 6 over first 5 cycles.**

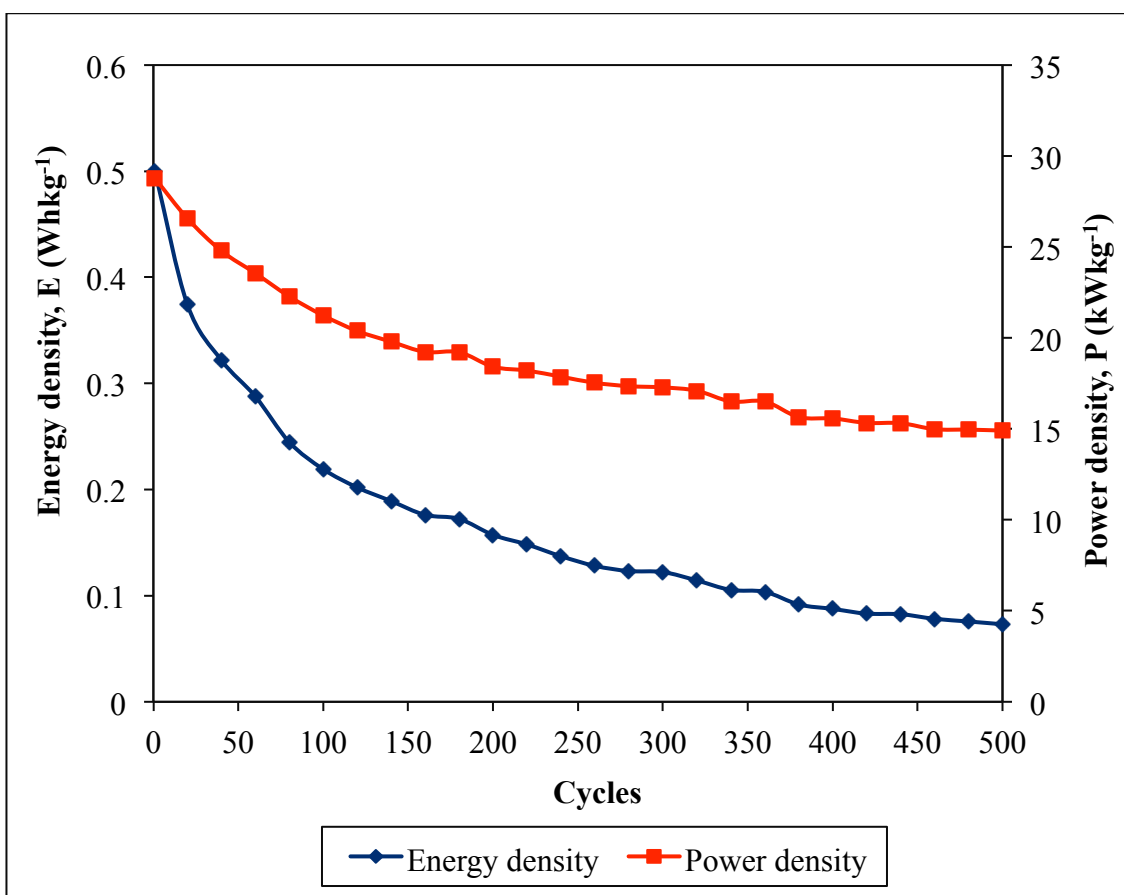
Figure 4.12 explains clearly the polymer electrolyte depletion phenomenon. As the cycle number increases, the internal resistance at both the charging and discharging increase with the cycle number. This is because ions would rather be paired up rather than be transported to the electrode electrolyte interface. This causes polymer electrolyte depletion, which results in increased internal resistance (Pandey et al., 2010).

It is important to determine the stability of the electrochemical cell upon charging and discharging processes. This is deemed as the cyclability test. Specific

capacitance, energy density and power density of EDLC using PVA 4 is depicted in Figures 4.13 and 4.14. For PVA 4, at the start of the cycle, the specific discharge capacity is  $6.84 \text{ Fg}^{-1}$ . The energy density and power density are both  $0.50 \text{ Wh kg}^{-1}$  and  $57.58 \text{ kW kg}^{-1}$ , respectively. However after 500 cycles, we do see a significant drop in all three parameters, with specific discharge capacity becoming  $4.31 \text{ Fg}^{-1}$ , energy density and power density both becoming  $0.10 \text{ Wh kg}^{-1}$  and  $33.01 \text{ kW kg}^{-1}$ .



**Figure 4.13: Specific capacitance of EDLC containing PVA 4 over 500 cycles.**



**Figure 4.14: Energy density and power density of EDLC containing PVA 4 over 500 cycles.**

PVA 6 as the highest conducting polymer electrolyte showed better performance when compared with PVA 4. At the beginning of the charge and discharge cycle, the EDLC shows a specific capacitance of  $45.25 \text{ Fg}^{-1}$ , energy density of  $4.00 \text{ Wh kg}^{-1}$  and power density of  $61.33 \text{ kW kg}^{-1}$ . These values are in line with the positive results obtained from CV. From the 1<sup>st</sup> to the 20<sup>th</sup> cycle, an abrupt drop is observed in the capacitance and energy density. The specific capacitance drops to  $31.40 \text{ Fg}^{-1}$  and the energy density decreases to  $2.22 \text{ Wh kg}^{-1}$ . The power density decreases but still maintains a high at  $54.85 \text{ kW kg}^{-1}$ . After the 20<sup>th</sup> cycle, the capacitance, energy density and power density decrease at a slow rate and at the 500<sup>th</sup> cycle the capacitance, energy density and power density are  $14.25 \text{ Fg}^{-1}$ ,  $0.48 \text{ Wh kg}^{-1}$  and  $37.87 \text{ kW kg}^{-1}$ , respectively.

The decrease in the energy density and power density value determining the electrochemical properties of the EDLC over the cycle number can be attributed to electrolyte depletion. As cycle number increases, the number density of the charge carriers decreases. Another noteworthy reason is the formation of ion aggregates. This can mean that the ions are paired up upon charging and discharging. These ion pairs can block the ion migration in the polymer electrolyte, which will affect ion adsorption into the pores of carbon electrodes. This in turn would reduce the ions, which accumulate at the electrode-electrolyte interface. As a result, capacitance of the EDLC decreases which may be also common factor in commercially available EDLCs and previously reported work by other researchers (Liew et al., 2014)

It is clear from both Figure 4.15 and 4.16 that the highest ionic conducting polymer electrolyte has maintained its efficient performance from the start of the study. Although PVA 4 showed the second highest ionic conductivity, its use in an electrochemical device may not be encouraged due to its stability over time when compared with PVA 6.

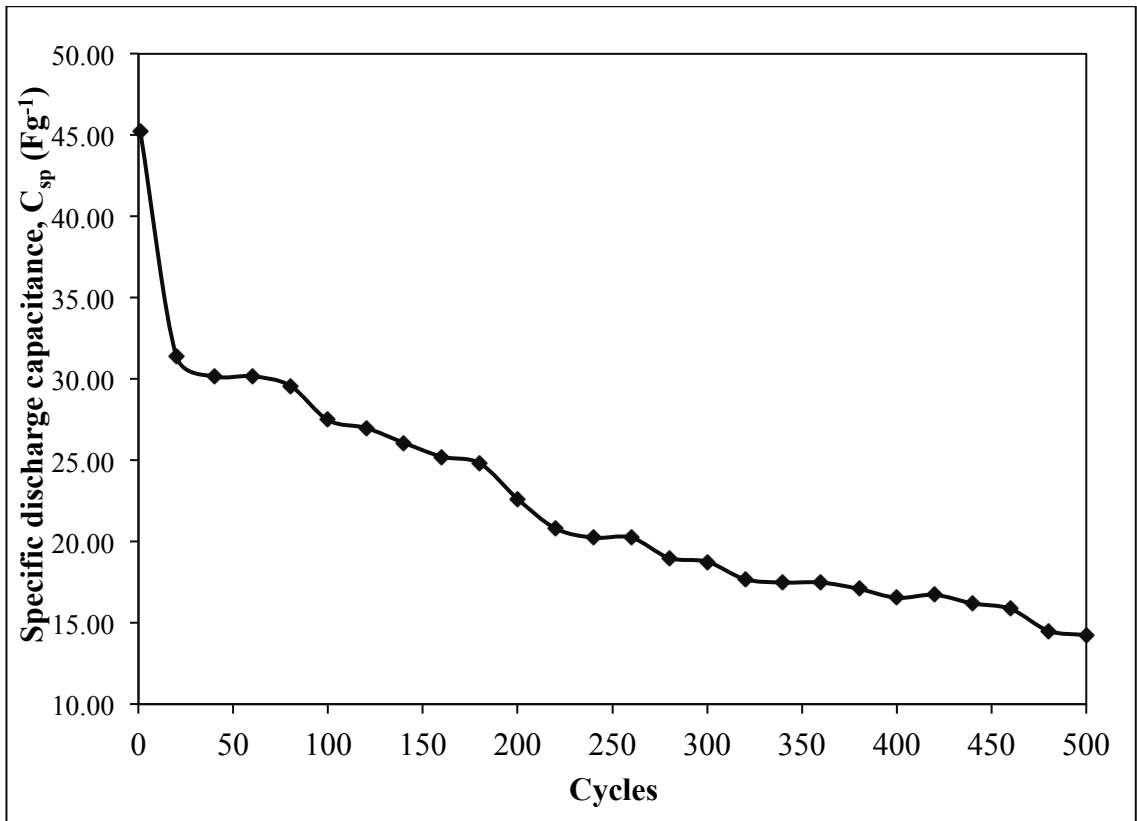


Figure 4.15: Specific capacitance of EDLC containing PVA 6 over 500 cycles.

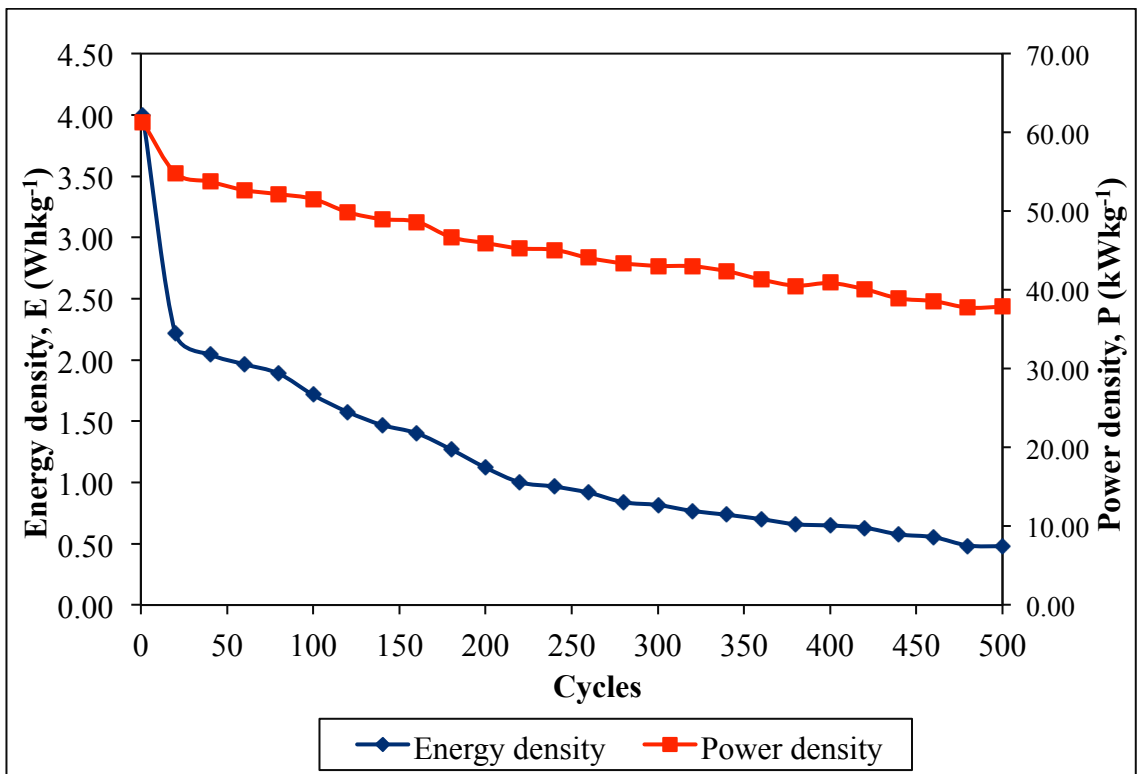


Figure 4.16: Energy density and power density of EDLC containing PVA 6 over 500 cycles.

## 4.8 Summary

The PVA-  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ - BmImBr based polymer electrolytes were successfully prepared and fabricated into working EDLCs. The PVA based polymer electrolytes were treated as biodegradable polymer electrolytes due to the biodegradable nature of the polymer and the use of ionic liquid. The highest ionic conductivity was obtained for the salt added polymer electrolyte doped with 60 wt% BmImBr with a value of  $(1.64 \pm 0.01) \times 10^{-3} \text{ Scm}^{-1}$ . Temperature dependent ionic conductivity confirmed that the polymer electrolyte samples incorporated with BmImBr were all found to obey VTF rule. DSC studies were done to show that glass transition temperature,  $T_g$ , was the lowest for the sample PVA 6, which correlated with the highest ionic conductivity obtained by the sample. This lowest  $T_g$  also evidenced the transition of the polymer electrolyte to the amorphous phase. Through XRD technique, complexation occurrence between PVA- $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ -BmImBr was confirmed. XRD results also show that the amorphous nature of the polymer electrolytes is increased with the incorporation of BmImBr. Upon fabrication of the device, it was subjected to CV and GCD and once again PVA 6 was found to perform well. The EDLC prepared with PVA 6 shows a specific capacitance of  $45.25 \text{ Fg}^{-1}$ , energy density of  $4.00 \text{ Wh kg}^{-1}$  and power density of  $61.33 \text{ kW kg}^{-1}$ .



## CHAPTER 5: RESULTS AND DISCUSSION ON POLY (ETHYL METHACRYLATE) (PEMA) BASED SYSTEM

This chapter discusses the results obtained for the poly (ethyl methacrylate), which is abbreviated as PEMA, Magnesium triflate salt ( $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ ) and the ionic liquid, 1-butyl-3-methylimidazolium bromide (BmImBr) system which was experimented as the non- biodegradable based polymer electrolyte system in this study. Acrylate polymers can generally be classified as plastics. These polymers do not degrade in the environment hence making it non-biodegradable. This non-degradability may be due to the strong interatomic bonding in the molecules, which make it difficult for microbes to digest making decomposition difficult. Another example of the acrylate family is poly (methyl methacrylate) (PMMA), which is used as bone cement in the orthopaedic field.

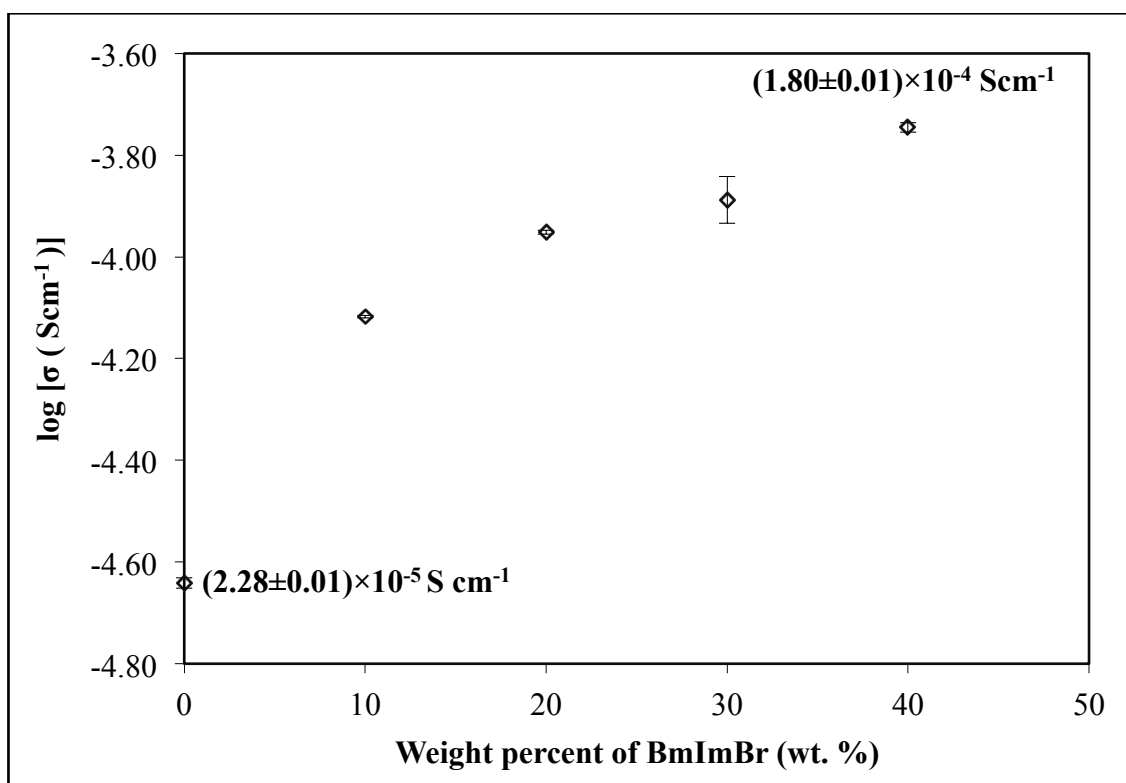
The first part of the study was the preparation of polymer electrolyte films with magnesium triflate salt and then with varied weight % of BmImBr. Ionic conductivity of these prepared polymer electrolytes were carried out at room temperature. Following that, temperature dependent ionic conductivity was done to investigate its ionic mobility mechanism from ambient temperature to 100°C. Differential Scanning Calorimetry was performed to determine the glass transition temperature ( $T_g$ ) of the samples. Determination of  $T_g$  is very important especially for acrylate type polymers like PEMA, PMMA and poly (propyl methacrylate) (PPMA). This is due to the large pendant groups which limit how closely the polymer chains can pack together subsequently effecting the free volume. The more separate the chains are, the more easily they can move around which then increases the translational mobility. Theoretically, this should lower the  $T_g$ . This will then be followed by X-ray diffraction analysis to determine the

occurrence of complexation between PEMA,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  and BmImBr. The electrochemical potential window of these prepared polymer electrolytes was established using a CHI600D electrochemical analyser through the linear sweep voltammetry (LSV) method. The second part of the study was then initiated, which was the fabrication of electric double layer capacitor (EDLC) using the highest conducting non biodegradable polymer electrolyte based on PEMA and followed by device characterization.

### **5.1 Ionic Conductivity Study of Polymer Electrolytes**

Results illustrating the ionic conductivity of PEMA- $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ -BmImBr polymer electrolyte system with reference to the different weight percentage of 1-butyl-3-methylimidazolium bromide (BmImBr) at ambient temperature up to 40 wt % of BmImBr are as shown in Figure 5.1.

Three ionic conductivity data for each sample is averaged to obtain the plotted ionic conductivity value. The error bars for each ionic conductivity value is small, indicating a more certain average value. In addition, the standard deviation obtained for the room temperature ionic conductivity values were in the range of 0.009 – 0.04. These low standard deviation values further prove the reliability of the ionic conductivity values obtained.



**Figure 5.1: Ionic conductivity of PEMA based polymer electrolytes with different weight percentage of BmImBr**

Figure 1 shows that ionic conductivity of PEMA-Mg(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub> (70:30) is  $2.28 \times 10^{-5} \text{ Scm}^{-1}$ . The ionic conductivity of pure PEMA can normally be in the range of  $10^{-11} \text{ Scm}^{-1}$ . This increase in ionic conductivity upon addition of salt would be due to the increase in the number of charge carriers in the polymer electrolyte. Consequently, the increase in charge carriers would increase ion movement, thus increasing ionic conductivity.

Jayaraman et al. (2016) and co-workers carried out a study on PEMA blended with PVdF-co-HFP/PbTiO<sub>3</sub> matrix of (Poly (vinylidene fluoride) (PVdF) with 12 mol% of hexafluoropropylene (HFP) incorporated with lead titanate (IV) (PbTiO<sub>3</sub>) as filler and ethylene carbonate (EC) and dimethyl carbonate (DMC) as plasticizer with Lithium bis(perfluoroethanesulfonyl) imide (BETI) as salt and obtained ionic conductivity in the order of  $10^{-5} \text{ Scm}^{-1}$ . Han et al. (2002) in a study with PVC/PEMA blend incorporated

with  $\text{LiPF}_6$  and with plasticizers EC and PC obtained highest ionic conductivity in the range of  $10^{-4} \text{ Scm}^{-1}$ . PEMA blended PVdF-co-HFP system was investigated by Sivakumar et al. (2007) and co-workers who reported the maximum ionic conductivity of  $1.5 \times 10^{-4} \text{ Scm}^{-1}$ .

It is noteworthy that ionic conductivity increases upon addition of ionic liquid. This is seen in the difference in ionic conductivity between PEMA 0 and PEMA 1 whereby upon addition of 10 wt% ionic liquid, the ionic conductivity increased to  $(7.63 \pm 0.01) \times 10^{-5} \text{ Scm}^{-1}$ . The highest ionic conductivity achieved was  $(1.80 \pm 0.01) \times 10^{-4} \text{ Scm}^{-1}$  with addition of 40 wt % BmImBr. This effect could be attributed to the fact that doping with BmImBr increases the amorphous nature of the polymer electrolyte. In general, decreasing the crystallinity would make the polymer backbone more flexible thus promoting better ionic transport (Kumar et al., 2009). This abrupt increase in the ionic conductivity can also be attributed to the strong plasticizing effect of the BmImBr used, which softens the polymer backbone consequently increasing the flexibility of the polymer chain. Higher flexibility of polymer chains is known to improve mobility of polymer segments, which in turn, assists in ionic transport within polymer complexes. The ionic liquid also has the ability to weaken transient coordinative bonds in the crystalline region molecules, enabling them to become more flexible which leads to a higher degree of amorphousness in the polymer complexes.

Factors that contribute to high conductivity are concentration of amorphous region, concentration of mobile cations and additional transit sites (Han et al., 2002). This is unlike the crystalline region where the ions vibrate and are more static along the polymer backbone, disallowing ion movement and current generation. On the other hand, an amorphous region allows more mobility in the matrix. There would be more

mobile cations that can be transported with the current flow, making the sample more conductive.

The ionic liquid also assists in the dissociation of  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  due to the weakening Coloumbic force between anion and cation of the salt itself, being the  $\text{Mg}^{2+}$  and  $(\text{CF}_3\text{SO}_3)_2^-$  (Yahya & Arof, 2004). The weakening therefore enhances ionic dissociation which increases the total number of charge carriers and ultimately increases the ionic conductivity (Han et al., 2002; Sirisopanaporn et al., 2009). Increase in the number of charge carriers also mean more conducting sites for ion transportation. Ionic liquid also acts to reduce intermolecular interaction by interacting with the host polymer, and as a result increases the amorphous nature of the polymer electrolyte.

Previous work done by Sim et al., (2012) using PEMA blends with lithium salt found that the highest ionic conductivity obtained was  $2.87 \times 10^{-7} \text{ Scm}^{-1}$ . Upon addition of two different plasticizers, ethylene carbonate (EC) and propylene carbonate (PC), they found that the ionic conductivity increases to  $1.05 \times 10^{-4} \text{ Scm}^{-1}$  and  $1.46 \times 10^{-6} \text{ Scm}^{-1}$  respectively. This result can be similarized with the present study whereby the ionic liquid acts as a plasticizer. As in Sim's study whereby the plasticizer increases the amorphous nature of the polymer electrolyte, so it is with our study where the amorphous nature is increased using ionic liquid. This shows the importance of plasticizing effect, which makes the polymer backbone more flexible, encouraging ion segmental mobility, which increases ionic conductivity. In addition to that Sim and co-workers also reported the importance of mechanical stability of the films. As important as ionic conductivity is, the mechanical stability is also important especially when using the polymer electrolytes for device applications.

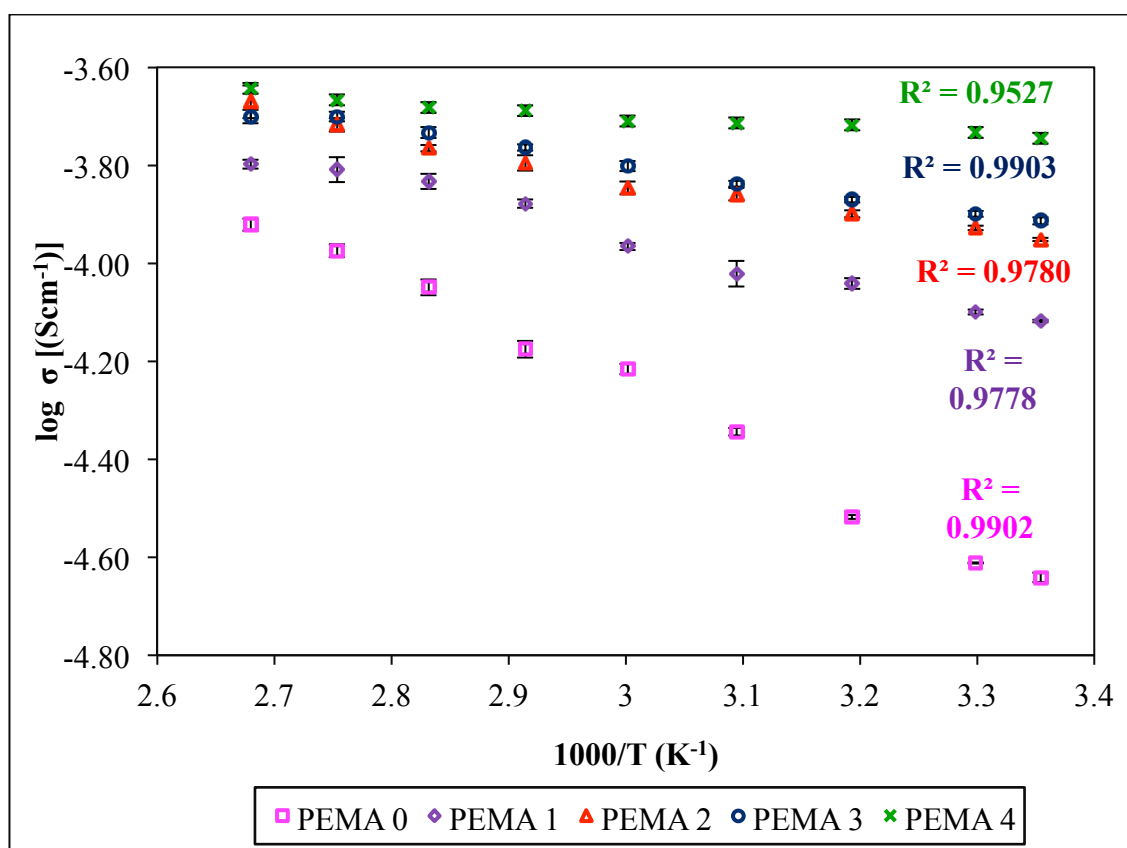
PEMA based polymer electrolytes incorporated with EC as plasticizer and lithium salt have also been found to exhibit conductivity in the  $10^{-4} \text{ Scm}^{-1}$  by Reiter et al. (2009). In a later study by Sim et al. (2014) using PEMA blends, Li salt and ionic liquid, the maximum ionic conductivity obtained was  $8.59 \times 10^{-5} \text{ Scm}^{-1}$ . These authors went one step further to experiment on how the ion mobility changed with increasing ionic liquid concentration. They found that the ion mobility increased and then decreased which they attributed to ion aggregates formed by the  $\text{Li}^+$  and  $\text{Tf}^-$  ions belonging to the salt and ionic liquid of the experiment. However, even when the ion mobility decreased the ionic conductivity was still on the increasing trend. Sim and co-workers (2014) suggested that this may be because most of the mobile ions are  $\text{Li}^+$  which are smaller compared to the  $\text{BMI}^+$  which is transported more easily in the polymer electrolyte matrix. As to where the  $\text{Li}^+$  ions came from, the researchers reported that the interaction between  $\text{BMI}^+$  cations with  $\text{Tf}^-$  anions of  $\text{LiTf}$  salt which then weakens the attraction between  $\text{Li}^+$  and  $\text{Tf}^-$  (Kang et al., 2006). In our study, the ionic liquid acts as a plasticizer. It increases the flexibility of polymer backbone, which breaks the transient partial bonding between mobile ions causing more charge carriers to migrate, resulting in higher conductivity. The imidazolium,  $\text{BmIm}^+$  is by itself a bulky cation, which causes bulk resistance and does not assist much in migration.

However, upon addition of more than 40 wt.% of  $\text{BmImBr}$ , the films become mechanically unstable. As important as ionic conductivity is, the mechanical stability of the film also determines their suitability in electrochemical devices (Sim et al., 2012). Due to these reasons, the film with 40 wt.% of  $\text{BmImBr}$  (PEMA 4) was selected as the optimum composition as it yielded the highest ionic conductivity of  $1.80 \pm 0.01 \times 10^{-4} \text{ Scm}^{-1}$  with acceptable mechanical stability.

## 5.2 Temperature Dependent Ionic Conductivity Studies

The polymer electrolytes are subjected to temperature dependent ionic conductivity testing to evaluate their ionic conduction mechanism. Figure 5.2 shows the log of ionic conductivity as a function of absolute temperature from ambient temperature to 100°C.

Three ionic conductivity data for each sample is averaged to obtain the plotted ionic conductivity value. The error bars calculated for each ionic conductivity value is small indicating a more definite average value. In addition, the standard deviation obtained for the ionic conductivity values were in the range of 0.003 – 0.02. A low standard deviation value indicates the accuracy of the ionic conductivity values and this is coupled with the small error bars as well.



**Figure 5.2: The temperature–dependent plot of PEMA based polymer electrolytes fitted with Arrhenius equation over wide temperature range**

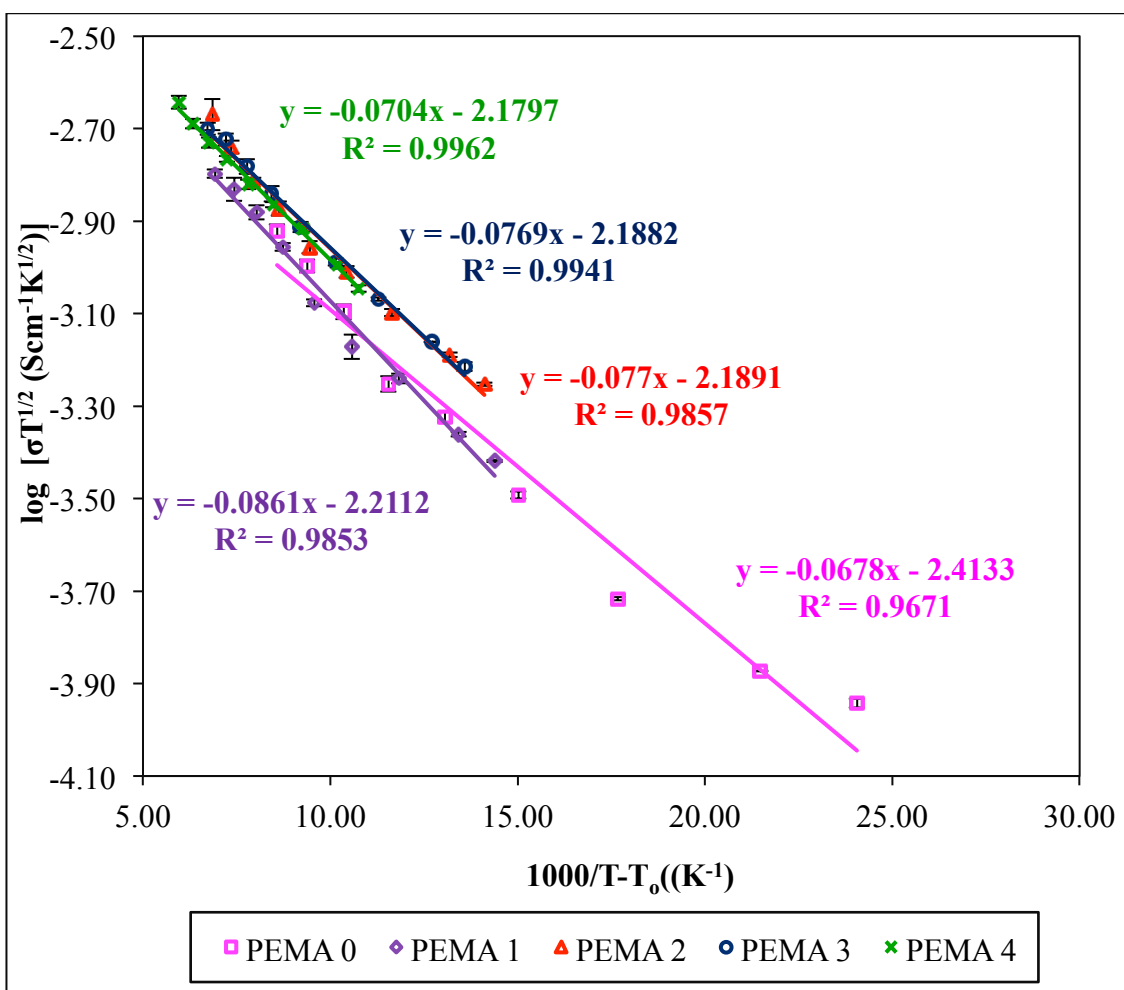
From the regression values obtained, it is found that PEMA 0 and PEMA 3 exhibit Arrhenius rules with a regression close to unity. Since the regression values of other BmImBr incorporated PEMA based polymer electrolytes deviate from unity, the plots are further fitted with Vogel-Tamman-Fulcher (VTF) equation and it is found that PEMA 1, PEMA 2 and PEMA 4 obey the VTF rule. In addition, PEMA 3 shows a regression value even closer to unity under VTF rule compared to Arrhenius rule. This is clearly depicted in Figure 5.3 where the regression values ( $R^2$ ) of the plots approach unity, which is tabulated using the equation below:

$$\sigma = A_o T^{-\frac{1}{2}} \exp\left(\frac{-B}{T - T_o}\right) = A_o T^{-\frac{1}{2}} \exp\left(\frac{-E_a/k_B}{T - T_o}\right) \quad (\text{Equation 5.1})$$

where  $A_o$  is the pre-exponential factor,  $B$  is determined from the gradient of the plot ( $K^{-1}$ ),  $E_a$  is pseudo-activation energy for conduction which is associated with polymer segmental mobility (eV),  $k_b$  is Boltzmann constant,  $T$  represents the absolute temperature (K) and  $T_o$  is the temperature where the free volume disappears and ionic mobility becomes zero (Marcilla et al., 2006).  $T_o$  which is the temperature at zero configuration therapy suggested as 50 K below the glass transition temperature ( $T_g$ ) which is determined from differential scanning calorimetry (DSC) findings (Jiang et al., 2006; Kim et al., 1999).

The regression values ( $R^2$ ) are tabulated in Table 5.1 and the results from the Equation 3 is expressed in Table 5.2.





**Figure 5.3: The temperature–dependent plot of PEMA based polymer electrolytes fitted with Vogel-Tamman-Fulcher (VTF) equation over wide temperature range**

From Figure 5.2 and Figure 5.3, it is clear that PEMA-  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  sample (designated as PEMA 0) obey the Arrhenius theory. All other ionic liquid-added PEMA based polymer electrolytes exhibit VTF behaviour, which is associated with the free volume model. This is seen from the regression value of the plots, which approach unity. This allows us to conclude that PEMA- $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ -BmImBr based polymer electrolytes follow VTF behaviour. Arrhenius behaviour is usually typical for crystalline materials, and VTF is more towards materials with amorphous nature.

VTF behaviour is different from Arrhenius behaviour in that it combines the ionic hopping mechanism with segmental mobility to facilitate ion movement. This is

based on free volume theory, which is prevalent in the amorphous phase of a polymer electrolyte. When temperature is increased, the polymer chains become more flexible due to high bond rotation. This backbone flexibility and bond rotation increases ion mobility and facilitates ionic transportation. Ion transport is enhanced due to the high ion mobility, which consequently creates more free volume. Thus, more vacant sites are created for charge carriers to be incorporated into the polymer electrolytes which would increase ion hopping, giving rise to an increase in ionic conductivity.

Upon carrying out the calculations using the non-linear least square technique, several significant results have been tabulated as below and discussed.

**Table 5.1: The obtained parameters from each VTF plot with the experimental glass transition temperature from DSC thermogram ( $T_0$ )**

<b>Sample</b>	<b>Regression value, <math>R^2</math></b>	<b>Glass transition temperature, <math>T_g</math> (K)</b>	<b>Ideal glass transition temperature, <math>T_0</math> (K)</b>
<b>PEMA 1</b>	0.99	278.65	228.65
<b>PEMA 2</b>	0.99	277.29	227.29
<b>PEMA 3</b>	0.99	274.52	224.52
<b>PEMA 4</b>	0.99	255.16	205.16

**Table 5.2: The obtained parameters from each VTF plot ( $A_o$ ,  $B$  and  $E_a$ )**

Sample	Log $A_o$	Pre-exponential constant, $A_o$ ( $\text{mScm}^{-1} \text{K}^{1/2}$ )	Gradient of the plot, $B$ ( $\text{K}^{-1}$ )	Pseudo-activation energy, $E_a$ (meV)
<b>PEMA 1</b>	-2.2112	$6.15 \times 10^{-3}$	0.0861	7.42
<b>PEMA 2</b>	-2.1891	$6.47 \times 10^{-3}$	0.077	6.64
<b>PEMA 3</b>	-2.1882	$6.48 \times 10^{-3}$	0.0769	6.63
<b>PEMA 4</b>	-2.1791	$6.62 \times 10^{-3}$	0.0704	6.07

To reiterate what has been mentioned earlier, PEMA 4 showed the highest conductivity. The parameters tabulated in Table 5.1 and 5.2 provide the evidence.  $A_o$  is the pre-exponential constant which is proportional to the number of charge carriers and  $B$  is related to the rate which the viscosity changes with temperature (Pandey et al., 2011). It is obvious from the  $A_o$  values that PEMA 4 shows the highest number of charge carriers. This result correlates with the sample's highest ionic conductivity. The easiest interpretation would be that PEMA 4 has the most charge carriers, which enhances its diffusive ability compared with the ionic liquid added polymer electrolyte samples. In order to confirm the deductions, the pseudo-activation energy,  $E_a$  of each sample was calculated. As expected, PEMA 4 was found to have the lowest pseudo-activation energy. This may be due to the flexibility of the polymer backbone added with the higher segmental mobility, which allows the ions to move or hop easily, hence requiring lower activation energy.

On the other hand, upon observing the values for PEMA 2, the lowest conducting sample, it was shown to exhibit the lowest number of charge carriers as well. This explains its low ionic conductivity. In addition to that, the pseudo-activation

energy of PEMA 2 was also the highest in comparison, which correlates with its low ionic conductivity.

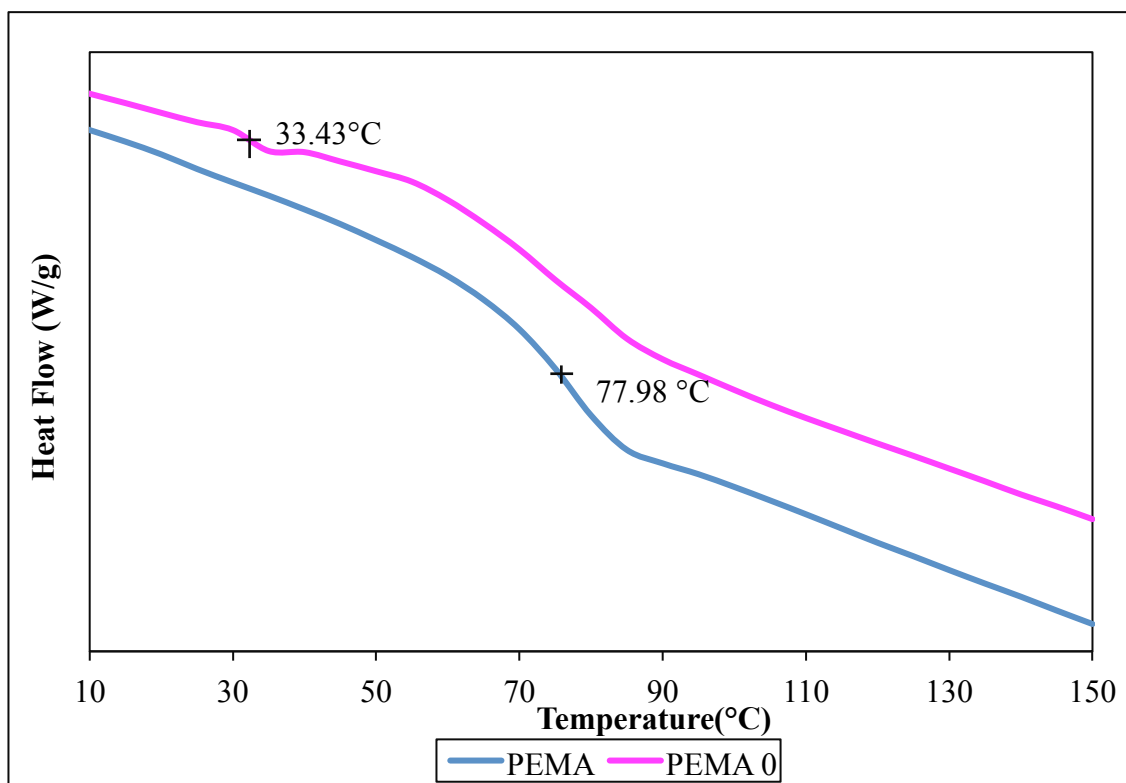
The pseudoactivation energy can be calculated from the slope of the VTF plot. Activation energy is the necessary energy required to move any chain segment for ion motion (Gong et al., 2012; Kumar & Suthanthiraraj, 2014). Decreased  $E_a$  gives rise to the highest ionic conductivity which is proven by PEMA 4 which is the highest conducting sample having lowest  $E_a$ . VTF theory surmises that the ionic conduction mechanism manifests the coupling effect where the ionic hopping mechanism is coupled with higher polymer segmental mobility in an amorphous phase (Ibrahim et al., 2012; Syahidah & Majid, 2015). This rule states that the increase in temperature would enhance the flexibility of the polymer chains due to the increase in thermal oscillation, which gives rise to faster movement of the polymer segments. Segmental mobility is hence improved with these flexible chains. Due to the high mobility of polymer segments, charge carriers can transport ions from one site to another, making the polymer electrolyte more conductive (Marcilla et al., 2006).

Previous reports have also mentioned that the increase in temperature promotes salt and ion dissociation, allowing more free ions to move around. In that study, the PEMA blends were found to obey VTF behaviour which the researcher attributed to the amorphous nature of the polymer electrolyte (Sim et al., 2012).

### **5.3 Differential Scanning Calorimetry (DSC)**

DSC is a tool necessary to define the glass transition temperature ( $T_g$ ) of the sample, which is the temperature when the polymer electrolyte undergoes phase transition from crystalline (hard, glassy) to a soft rubbery or amorphous state. It is

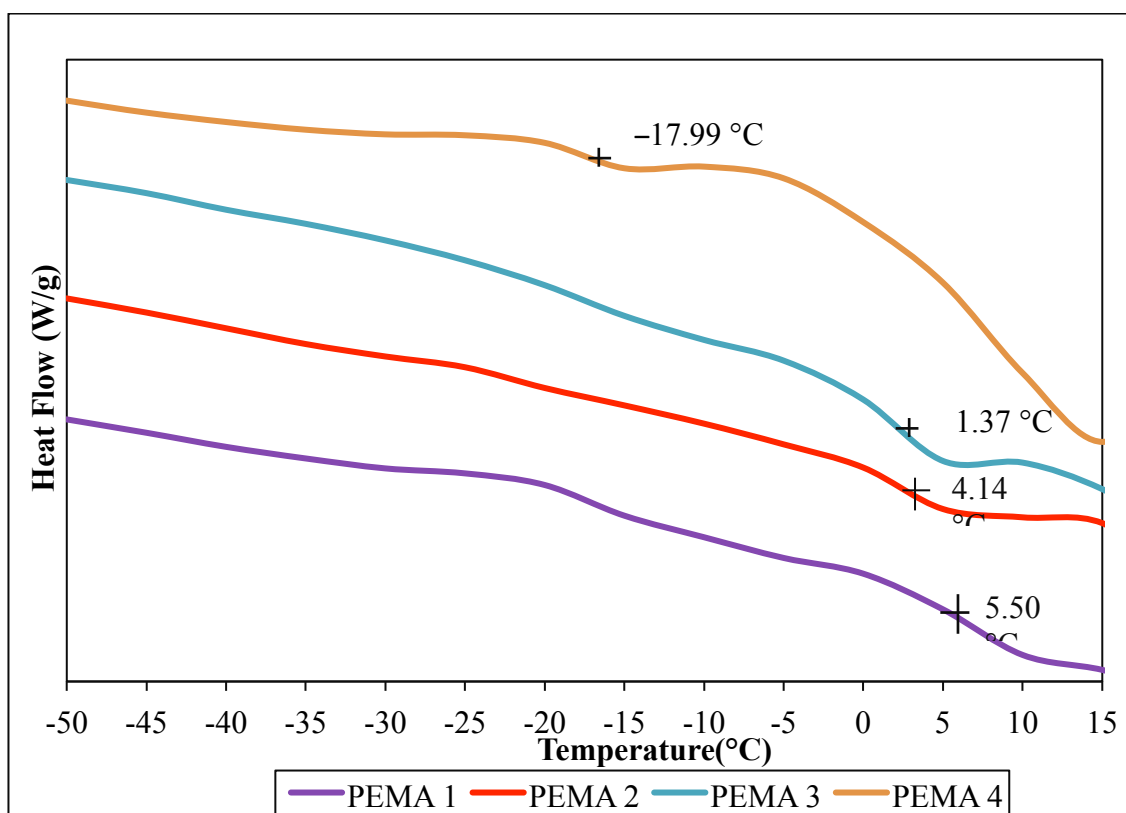
generally known that a polymer electrolyte with higher amorphous region would exhibit lower  $T_g$ . Hence, the lower the  $T_g$ , the more flexible the polymer backbone which in turn gives rise to higher conductivity due to the high mobility of ions.



**Figure 5.4 : DSC thermogram of pure PEMA and salt added PEMA.**

DSC curves of pure PEMA and Magnesium Triflate added PEMA in the temperature range between 10°C to 150°C is depicted in Figure 5.4. The DSC in Figure 5.4 shows that pure PEMA has a  $T_g$  of 77.98 °C and salt added PEMA has a  $T_g$  of 33.43 °C. In addition, no crystalline peaks were observed in the studied temperature range. This is an indication that the reaction is taking place in the amorphous region of the polymer electrolyte. This proves that the addition of  $Mg(CF_3SO_3)_2$  salt has reduced crystallinity of the polymer making it amorphous. Upon addition of salt, the backbone chains become more flexible, hence the lower  $T_g$  value for the salt added polymer

electrolyte. The decrease  $T_g$  also verifies the interaction existing between the salt and polymer.



**Figure 5.5 : DSC thermograms of ionic liquid added PEMA**

Figure 5.5 depicts the DSC thermogram for PEMA- $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ -BmImBr polymer electrolytes with different wt% of BmImBr at temperature range between  $-50^\circ\text{C}$  to  $15^\circ\text{C}$ . Upon addition of ionic liquid, in Figure 5.5, the  $T_g$  of PEMA 4, which is the highest conducting polymer electrolyte, has reduced dramatically to  $-17.99^\circ\text{C}$ . This significant reduction ascertains the plasticizing effect of the ionic liquid, which reduces the  $T_g$ . The consequence is a disrupted crystalline structure of the PEMA making it more amorphous (Jarosik et al., 2006). This improves flexibility of the polymer chain hence stimulates higher segmental mobility that is proven through the portrayal of high ionic conductivity. According to the VTF theory, the ion hopping is combined with high segmental mobility of the polymer chains, which takes place more conveniently in

the amorphous phase. It is this amorphous nature that is seen through the low  $T_g$  that we obtained experimentally, which explains the rubbery nature of the polymer.

When plasticizers or ionic liquids, which give plasticizing effect, are added into polymers, viscosity of the polymer-plasticizer mix would decrease. This is due to the replacement of polymer-polymer hydrogen bonding with polymer-plasticizer bonds. According to free volume theory, when molecules such as plasticizers are added into the polymer, the free volume available to the polymer chain increases and this in turn causes the decrease of the glass transition temperature.

#### **5.4 X-Ray diffraction**

Figure 5.6 presents the XRD patterns of PEMA,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  salt, BmImBr ionic liquid and PEMA-4. There was no sharp and well-defined peak observed for PEMA, except two broader peaks in range of  $10^\circ$  to  $22^\circ$  at  $2\theta$  value. This confirmed the presence of amorphous regions and non-crystalline structures (Fares, 2012).  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  demonstrated sharp and well define crystalline peaks at  $2\theta$  value of  $18^\circ$ ,  $22^\circ$ ,  $23.8^\circ$ ,  $29^\circ$ ,  $30^\circ$  and  $42.9^\circ$  which reveals its high crystalline nature. Upon incorporation of 40 wt. % of BmImBr ionic liquid and  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  salt, the characteristic peaks of  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  salt disappear in PEMA-4 polymer complex. This affirms the successful complexation of salt an ionic liquid in the host polymer.

Addition of ionic liquid reduces the intermolecular interaction of the polymer chains. This causes an increase in the amorphous region. The ionic conduction occurs in the amorphous phase of the polymer electrolyte due to the flexibility of the polymer backbone chains. In addition, the absence of additional peaks in the XRD also proves

that complexation has taken place and that the materials involved in the polymer electrolyte are compatible.

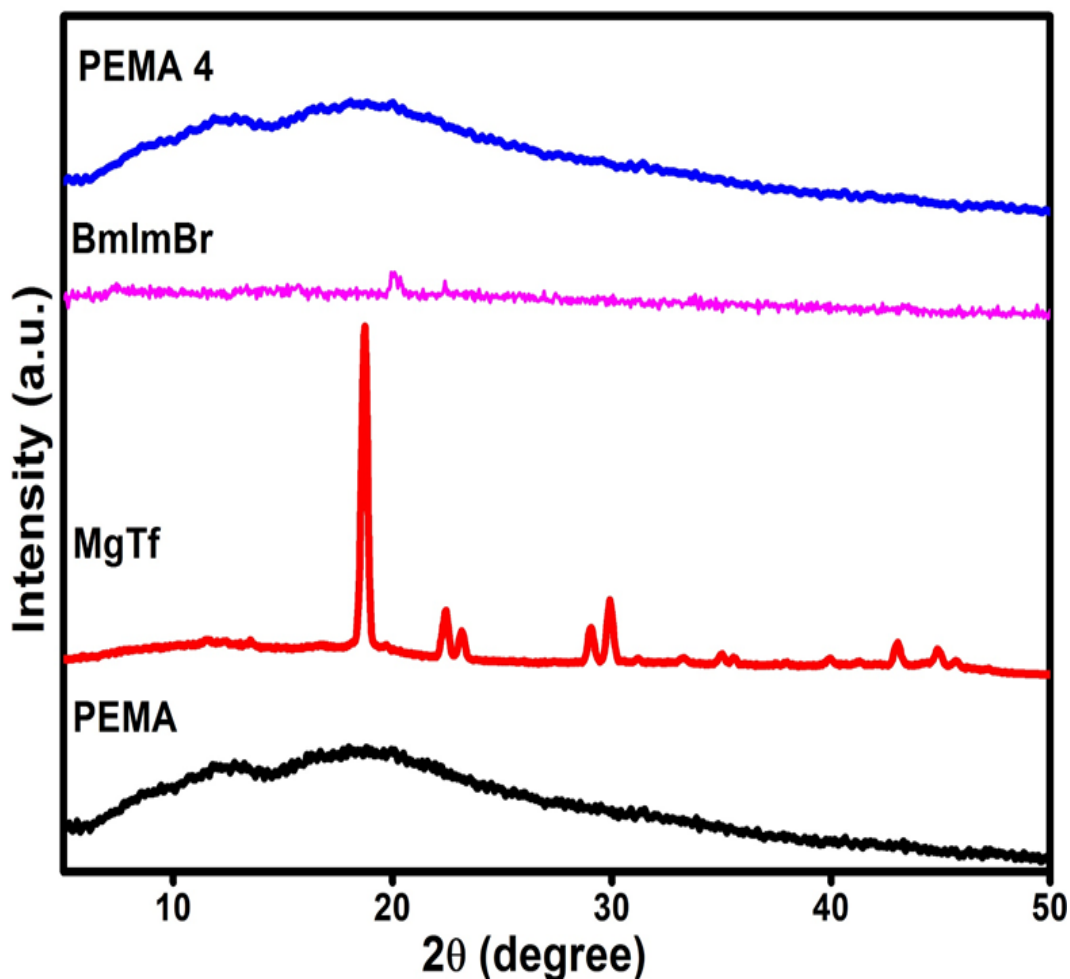


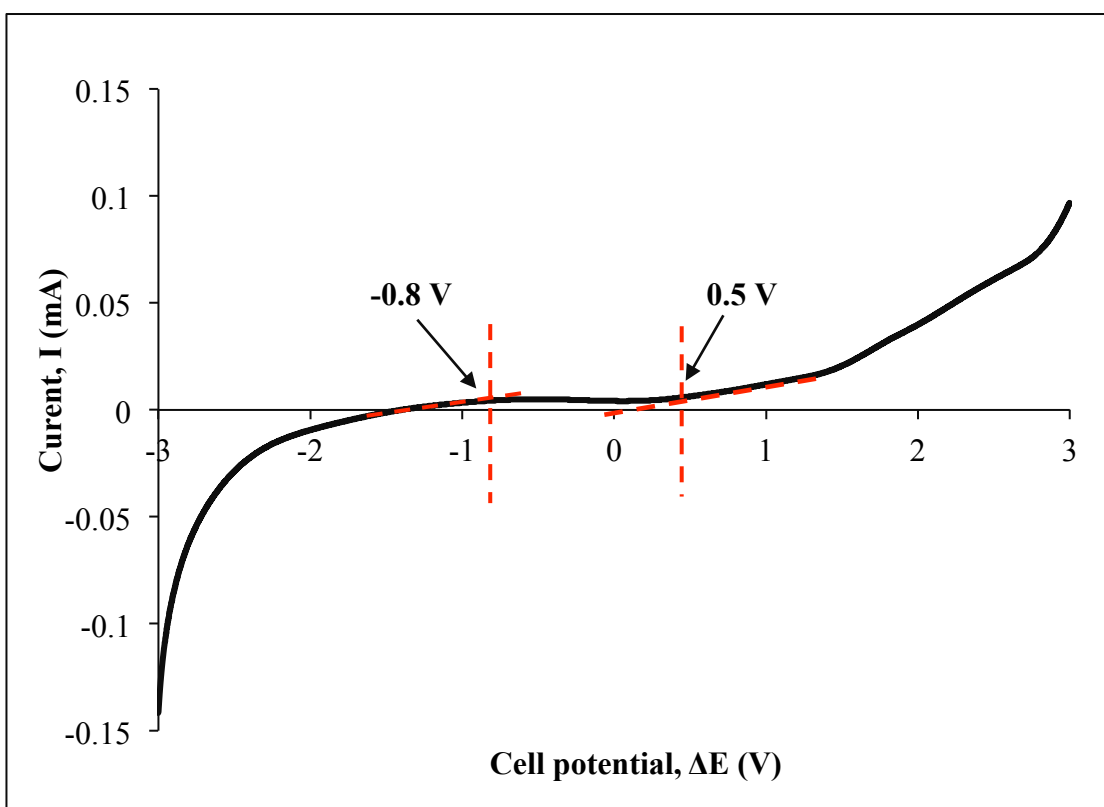
Figure 5.6: XRD patterns PEMA,  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ , BmImBr, PEMA 4

### 5.5 Linear Sweep Voltammetry (LSV)

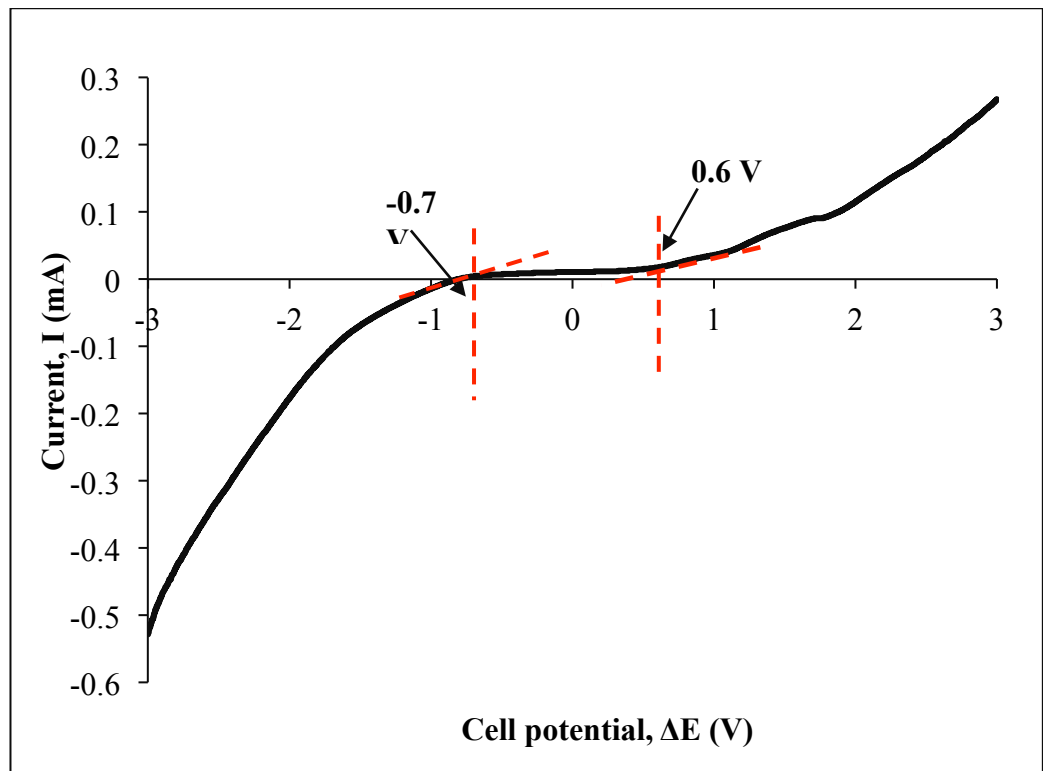
Linear Sweep Voltammetry is important to determine the maximum operational potential for a polymer electrolyte. Figures 5.7(a) and (b) represent the potential window of PEMA 0 and PEMA 4 respectively.



The LSV of PEMA-Mg(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub>, was 1.3 V, where the cathodic and anodic potential were detected at - 0.8 V and 0.5 V as can be seen from Figure 5.7(a). Upon impregnation with 40 wt% ionic liquid as seen in Figure 5.7(b), the operational potential window is detected at - 0.7 V and 0.6 V which is actually 1.3 V. LSV results shown in Figures 5.7 (a) and (b) for PEMA 0 and PEMA 4 indicate that the electrochemical potential window of the ionic liquid free and ionic liquid added polymer electrolyte remain the same at 1.3 V. LSV is important as it determines the “safe” maximum operating temperature of that particular electrochemical device and how long the device stays stable (Tiruye et al., 2016).



**Figure 5.7(a) : LSV response of PEMA 0.**



**Figure 5.7(b) : LSV response of PEMA 4.**

From both the LSV results, it was noted that although the PEMA 4 showed the highest ionic conductivity, in terms of electrochemical potential window, the addition of ionic liquid did not cause an improvement.

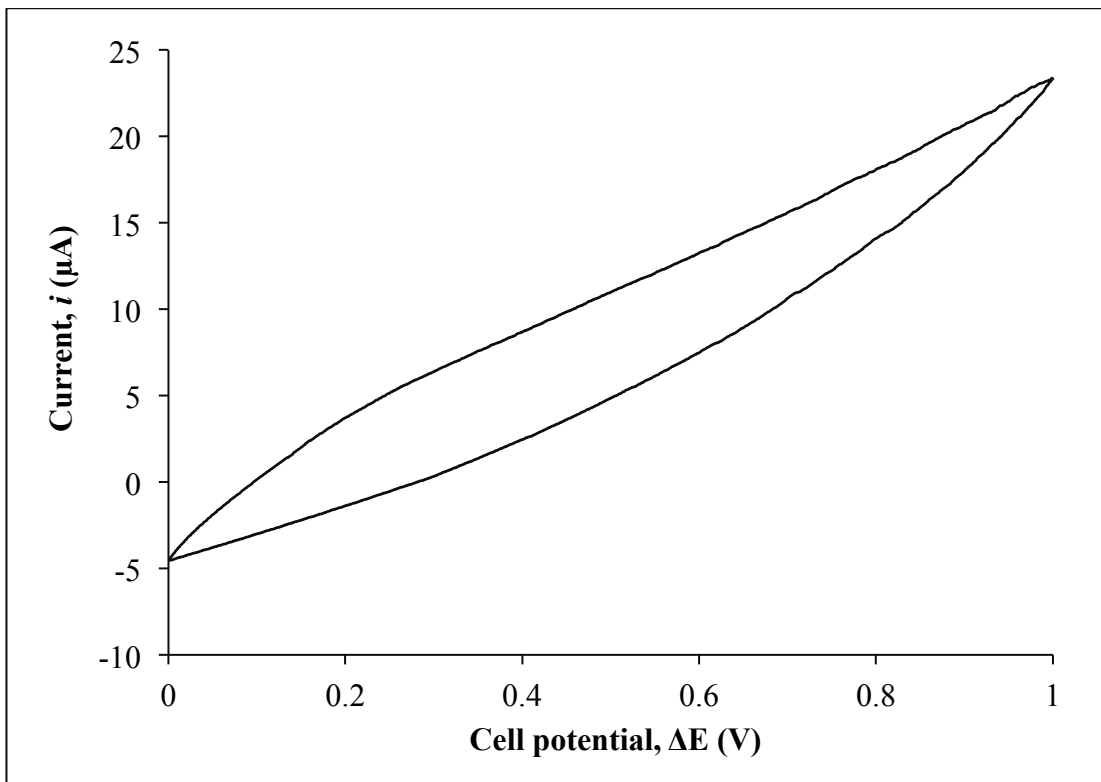
## 5.6 Cyclic Voltammetry (CV)

The CV curve of PEMA 0 and PEMA 4 are depicted in Figure 5.8 and 5.9 respectively. Both display a leaf-like shape with a specific capacitance of  $1.99 \text{ Fg}^{-1}$  for the ionic liquid free PEMA and  $7.34 \text{ Fg}^{-1}$  for the polymer electrolyte with 40 wt% PEMA.

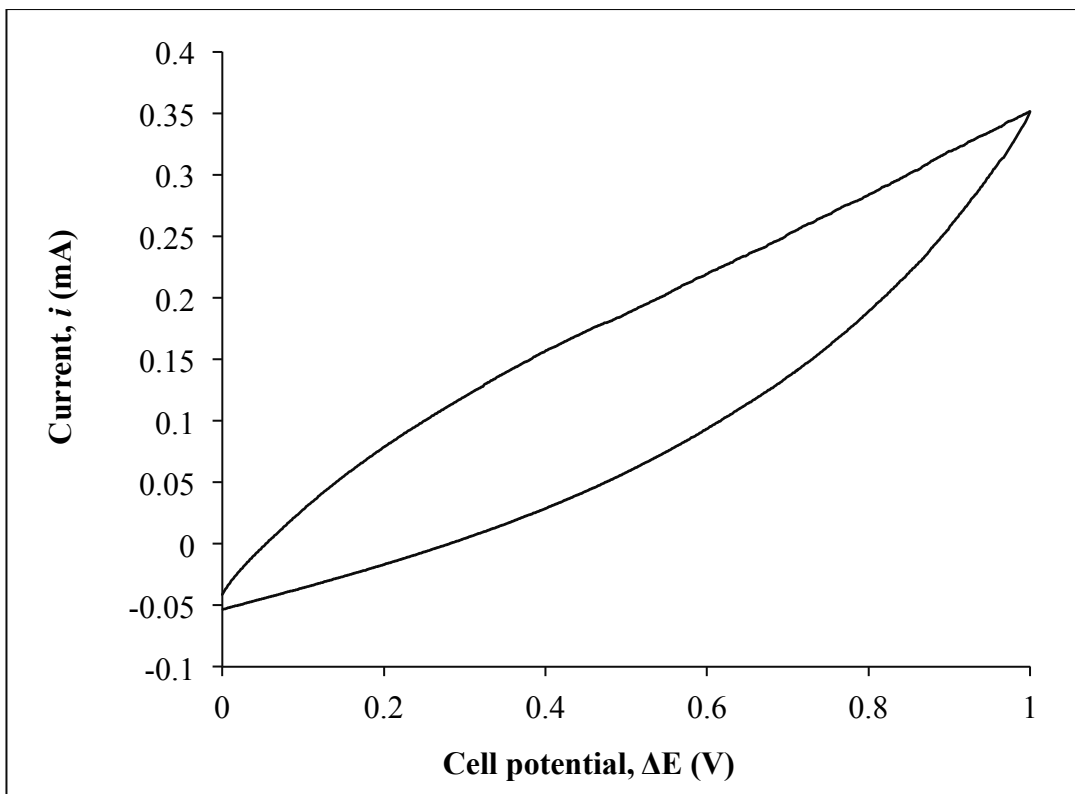
The absence of an ideal rectangular shape implies the poor utilization of solid polymer electrolyte in the EDLC which can be attributed to poor interfacial contact between electrode and electrolyte (Latham et al., 2002).

The comparatively low specific capacitance of PEMA 0 when compared with the other PEMA-Mg(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub>-BmImBr is also attributed to the poor ionic conductivity of the polymer electrolyte and poor interfacial contact between the electrode-electrolyte. PEMA 4 shows a comparatively higher specific capacitance, which may be due to the high mobility of ions in the polymer matrix of PEMA 4. This phenomenon is explained through the free ions that drift from one electrode to the other and get absorbed into the ion pores. This forms a charge accumulation at the electrode-electrolyte surface of the double layer due to increased ion mobility and ion content which in turn, gives rise to higher capacitance because of the excessive accumulation of charge carriers at the boundary.

Although the shape of the voltammogram does not portray the performance of the device, the comparatively higher value of specific capacitance showed that the ionic liquid incorporated polymer electrolyte is better in terms of performance. However, it is not high enough for device usage. This higher value of specific capacitance can be attributed to the presence of ionic liquid, which facilitated the ion migration within the polymer matrix. This high number of ions induces high mobility causing diffusion of ions into the pores of the porous electrode. The ions then accumulate at the electrode-electrolyte boundary, which promotes a double layer formation, which then promote an increase in the energy storage capability of the EDLC.



**Figure 5.8 : CV curve of the ionic – liquid free EDLC**



**Figure 5.9 : CV curve of 40% ionic – liquid added EDLC (PEMA 4)**

## 5.7 Galvanostatic Charge Discharge (GCD)

Constant current charge-discharge is used to evaluate electrochemical storage and pattern of EDLC. The repetitive loop of charging and discharging is known as a cycle. The working voltage of an EDLC is attributed to the electrolyte breakdown voltage whereas electrode resistance and conductivity would affect its internal resistance and capacitance.

The attempt to carry out an evaluation of electrochemical storage capability of the fabricated EDLC using PEMA 4 did not yield any results. An error message was obtained throughout the procedure. This can be attributed to the poor interfacial contact between the electrode-electrolyte. When there is poor contact between the electrode-electrolyte, there is no possibility of charge storage. It was also clearly seen in the specific capacitance analysis that the capacitance value obtained was low.

It can be concluded that although a reasonable ionic conductivity was obtained for PEMA 4, its physical attributes prevented it from giving good performance when fabricated into a device.

## 5.8 Summary

The PEMA-  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$ - BmImBr based polymer electrolytes were successfully prepared and fabricated into working EDLCs. The PEMA based polymer electrolytes were non-biodegradable due to the nature of the polymer. The highest ionic conductivity was obtained for the salt added polymer electrolyte doped with 40 wt% BmImBr with a value of  $(1.80 \pm 0.01) \times 10^{-4} \text{ Scm}^{-1}$ . Above 40 wt%, the polymer film became mechanically unstable. Temperature dependent ionic conductivity confirmed that the polymer electrolyte samples incorporated with BmImBr were all found to obey

VTF rule. DSC studies revealed that the lowest glass transition temperature,  $T_g$ , was obtained for the sample PEMA 4, which had the highest ionic conductivity. This lowest  $T_g$  also evidenced the transition of the polymer electrolyte to the amorphous phase. XRD technique evidenced the occurrence of complexation between PEMA- $Mg(CF_3SO_3)_2$ -BmImBr. XRD results also showed that the amorphous nature of the polymer electrolytes is increased with the incorporation of BmImBr. Upon fabrication of EDLC, the device was subjected to CV. However, even the highest conducting polymer electrolyte, PEMA 4, failed to yield a rectangular plot for the CV which can be attributed to the poor interfacial contact between electrode and electrolyte.

## CHAPTER 6: RESULTS AND DISCUSSION - SURVEY

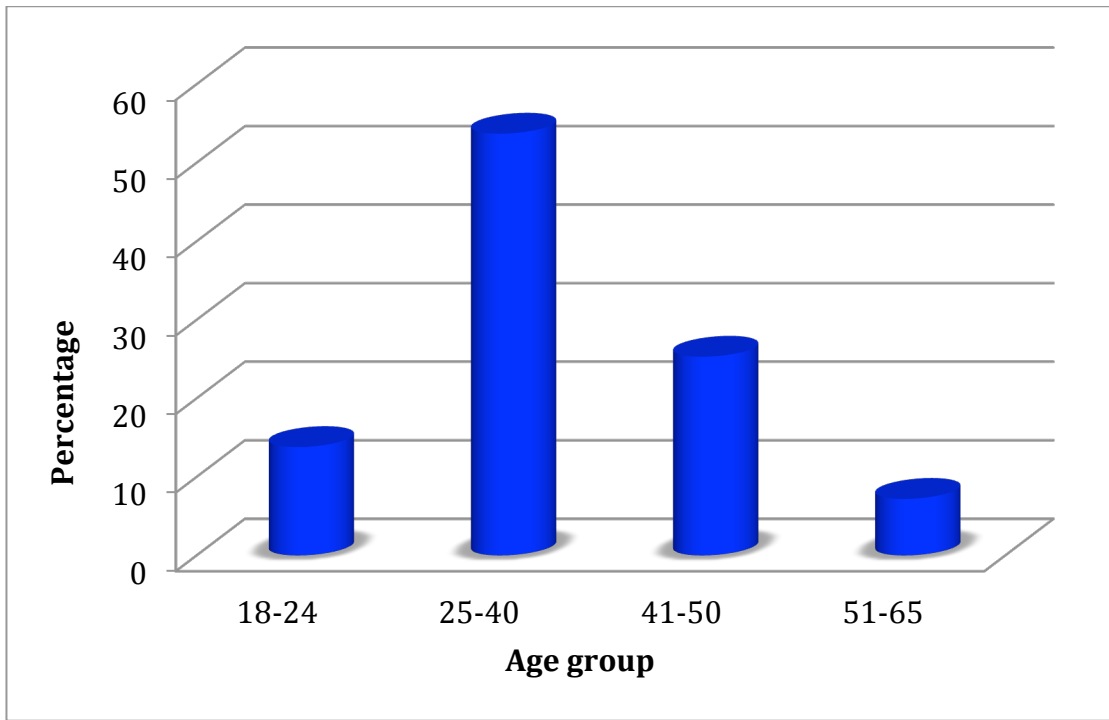
This chapter will evaluate the outcome of the survey data obtained. A description of the survey will be dissected which will be followed by the analysis using the Statistical Package for the Social Sciences (SPSS).

As the survey method used was an online survey, the total number of complete survey received was 391 responses which fulfils the minimum required number of samples as reported by Hair et al (2010).

### 6.1 Demographics

A wide array of demographic data was collected in order to understand the participants of the survey. This included gender, age, level of education, occupation, marital status and ethnicity. As our intention was to determine Malaysian adults willingness to pay for green cellphones, our population was mainly made up of working adults. This study intends to analyse the working community's willingness to pay for green electronics.

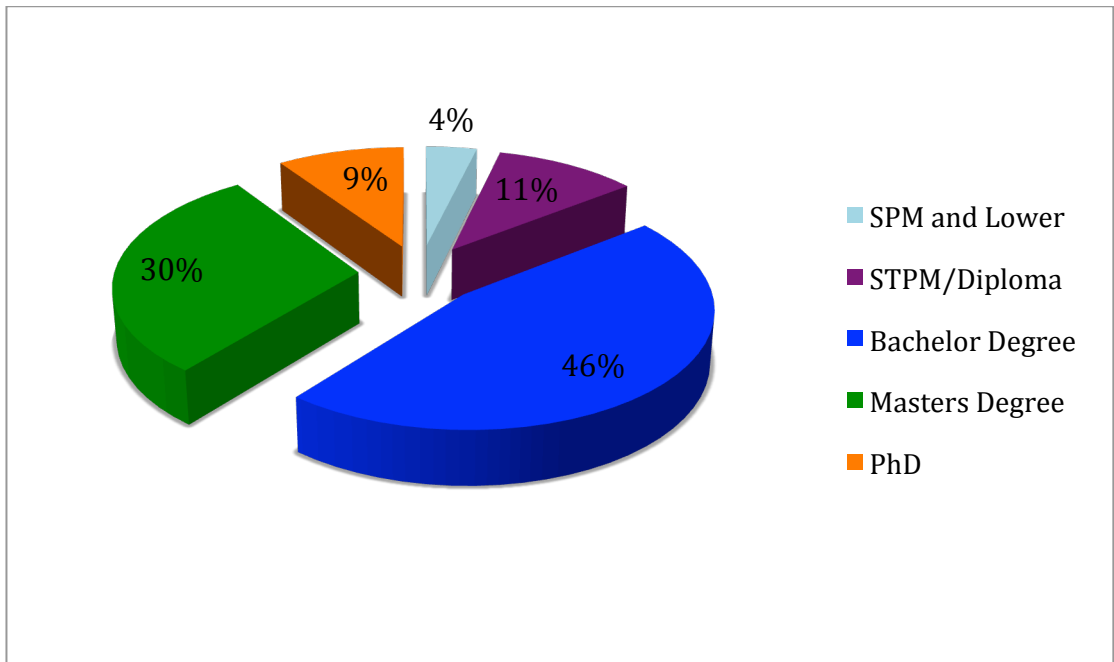
A total number of 239 female and 152 male respondents were obtained in the study. Our data was also made up of about 64 % married respondents.



**Figure 6.1: Age group of respondents**

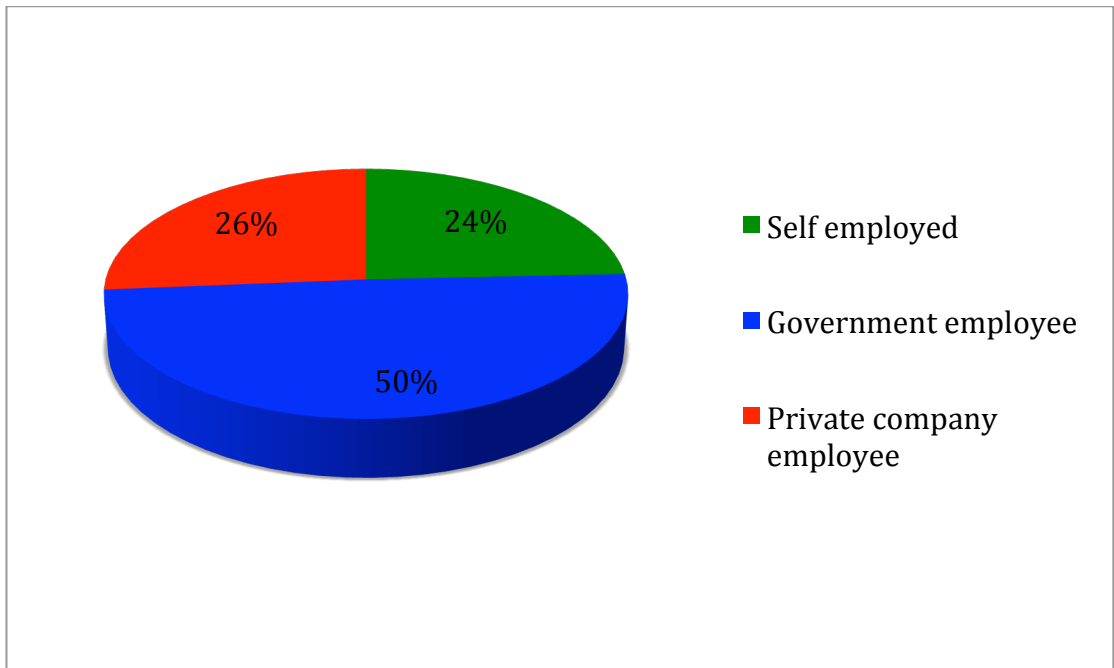
Figure 6.1 shows the age group of respondents obtained in our study. From the figure above, the highest age group was in the range of 25 – 40 which had 210 respondents, followed by 41 – 50 with 99 respondents. This age group can be considered the peak age group for working adults. The ages 18 – 24 had the third highest number, which would mainly comprise the young adults just embarking on their professional journey.





**Figure 6.2: Education level of respondents**

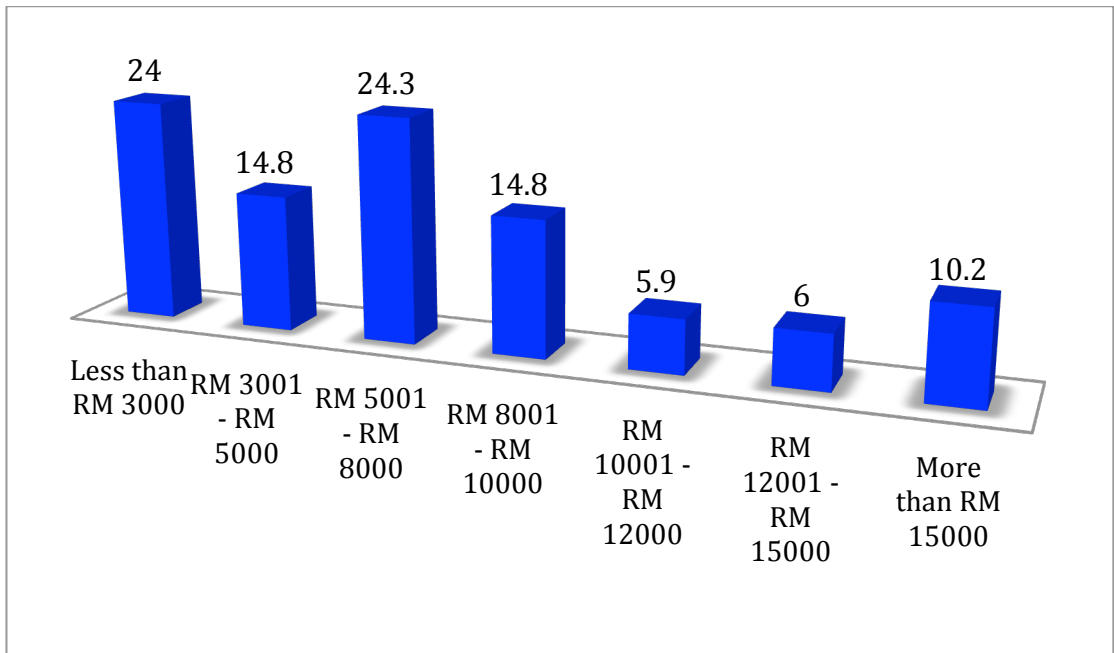
The chart in Figure 6.2 depicts education level of respondents. Interestingly, majority of the respondents, about 46 %, had a Bachelor’s degree. Those with a Masters degree followed this with a percentage of 30 %. The respondents clearly correlate with the demographic criteria, which is the current occupation. The minimum number was for lower than SPM. Education level was important to study the thinking and opinions of respondents when it came to buying green electronics.



**Figure 6.3: Occupation of respondents**

Figure 6.3 tabulates the job profile of respondents. Nearly 50 % of our respondents were government employees. Self employed and private company employees were nearly equal in amount. This result correlated with the level of education because government employees have the opportunity to pursue higher education while working which may be one of the reasons for the high number of Masters holders in our sample population. Thus, one can conclude, that even though most working adults are covered in the sample population, there is a clear dominance of government employees, reflecting the sampling technique chosen being convenience sampling.

Marital status was another demography surveyed to see if there was any relationship between marital status and willingness to pay. In our study, we had 64 % married respondents and 36 % single respondents.



**Figure 6.4: Household income of respondents**

An important information collected in this study was the monthly household income of respondents that is seen in Figure 6.4. The graph shows that 24 % had income of less than RM 3000 and between RM 5001 – RM 8000. This may correlate with the fresh graduates who have just started their job and those fresh diploma holders who newly started their jobs as well. The second group may be the ones who are more established in their career and have started earning nearly RM 8000.

The third and fourth highest earners were in the RM 3001 – RM 5000 and RM 8001 – RM 10000 categories. In general, it can be concluded that the respondents do have a reasonably high income. An objective of the study was also to investigate whether household income of respondents affects the consumers' willingness to pay for green electronics. In addition, the research also investigates whether the household income is proportional to the consumers' ability to pay for green electronics. A descriptive statistics summary of the tables above is tabulated in Table 1.

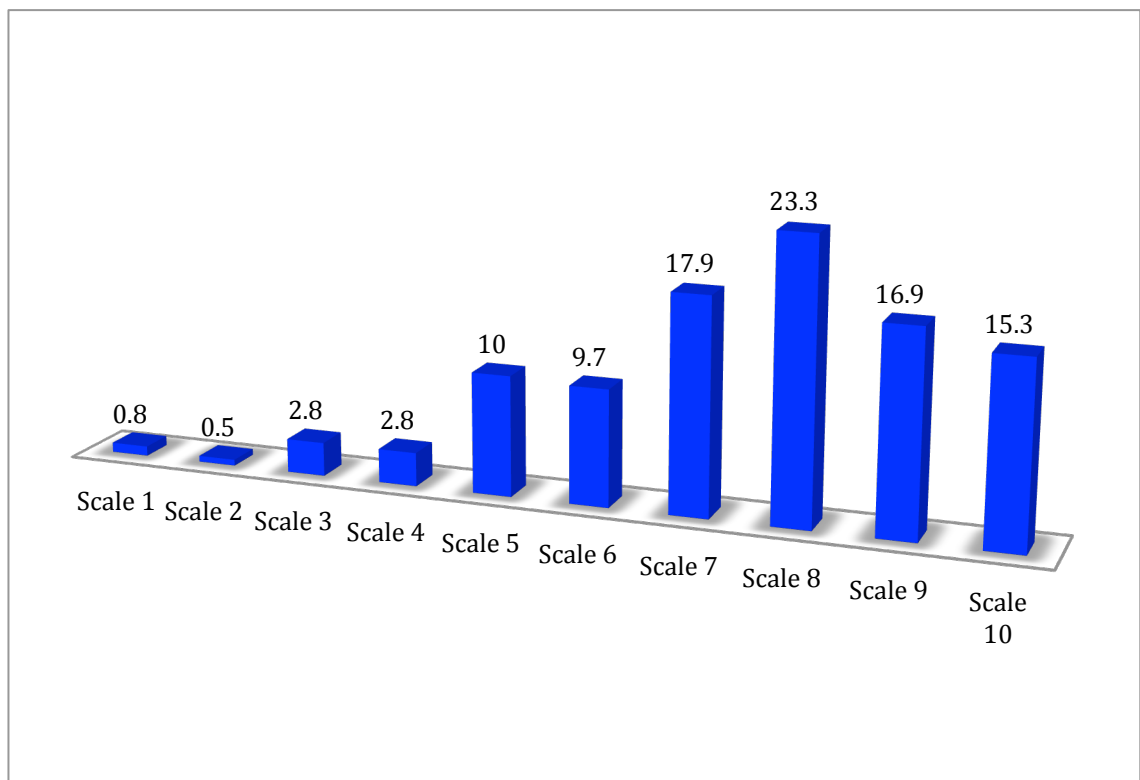
**Table 6.1: Descriptive Statistics**

	Frequency	(Percent)	Mean	Std. Dev
<b>Willing to Pay for Green Electronics</b>				
Yes	217			
No	174			
<b>Willingness to Pay at scale 1-10</b>			7.47	1.9
<b>Age Group</b>				
18-24	54	13.8		
25-40	210	53.7		
41-50	99	25.3		
51-65	28	7.2		
<b>Education Level</b>				
SPM and Lower	15	3.8		
STPM/Diploma	42	10.7		
Bachelor Degree	181	46.3		
Masters Degree	116	29.7		
PhD	37	9.5		
<b>Income</b>				
Less than RM 3000	94	24		
RM 3001 - RM 5000	58	14.8		
RM 5001 - RM 8000	95	24.3		
RM 8001 - RM 10000	58	14.8		
RM 10001 - RM 12000	24	5.9		
RM 12001 - RM 15000	22	6		
More than RM 15000	40	10.2		
<b>Occupation</b>				
Self employed	95	24.3		
Government employee	193	49.4		
Private company employee	103	26.3		
<b>Gender</b>				
Male	152	38.9		
Female	239	61.1		
<b>Marital Status</b>				
Married	247	63		
Single	144	37		
<b>Ethnicity</b>				
Malay	157	40.2		
Chinese	50	12.8		
Indian	162	41.4		
Sabah/Sarawak	5	1.3		
Others	17	4.3		

Ethnicity was asked as one of the demographic questions to investigate whether ethnicity was a dependent variable to willingness to pay for green electronics. In this study, the number of Malay and Indian respondents was nearly equal, with 40% for each. Chinese respondents made up 13% while the other 7% were made up of Others which comprised Sabahans and Sawarakians and other nationals who have made their home in Malaysia.

## 6.2 Willingness to Pay for Green Electronics

One of the questions asked in the survey was how consumers' would rank buying a "green" cellphone compared to other charitable acts and would they be willing to purchase a 'green cellphone. '1' indicates that they view buying a "green" cellphone much less favorably and 10 much more favorably than other causes. The results obtained were tabulated in Figure 6.5.

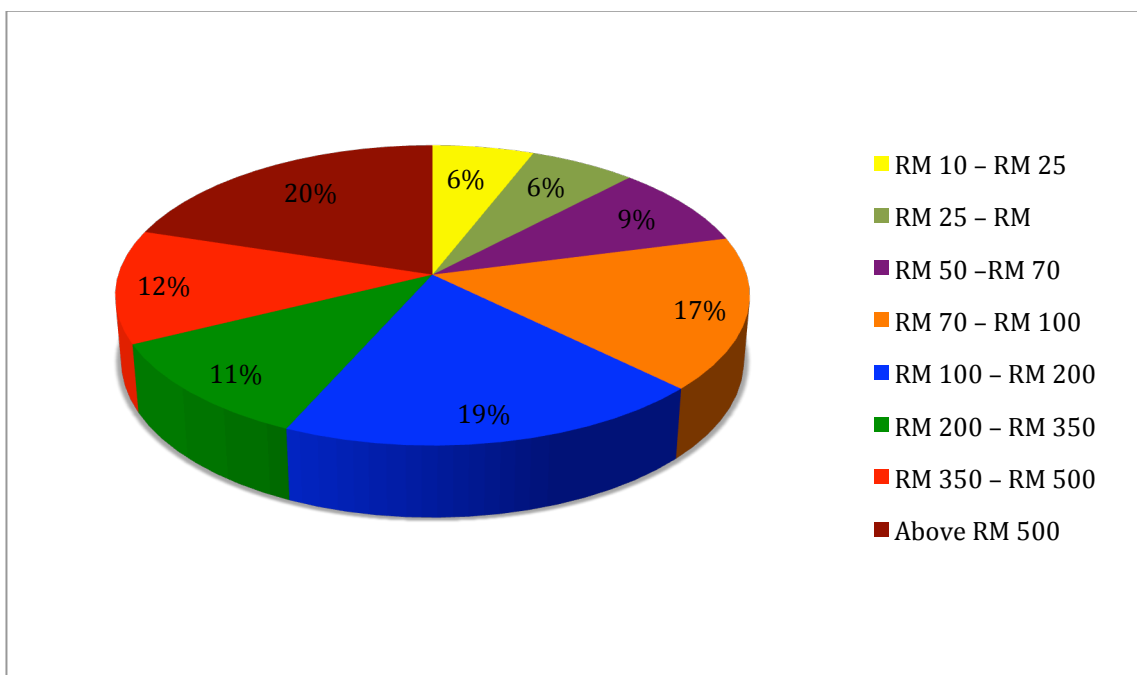


**Figure 6.5: Perceived Willingness to Pay for Green Electronics (%) compared to Charitable Acts**

From the above figure, it is clearly noted that more consumers' were willing to pay for green electronics compared to those who were not. The mean of ranking to purchase green electronics was calculated to be 7.47, hence 7 and below is recognized as not willing to pay [0] while 8,9,10 will be recognized as willing to pay [1]. This then provides the binary determinant as required for analysis by Logistic Regression.

Once the consumers' rate on how they would perceive willingness to pay for green electronics, they were asked this question:

“What is the highest amount, that your household would pay EXTRA ONLY ONCE for purchasing a “green” cellphone instead of a conventional cellphone (with exactly the same features and reliability)?”



**Figure 6.6: Amount Willing to Pay EXTRA only ONCE for a Green Cellphone**

Figure 6.6 depicts how much consumers are willing to pay ONCE to purchase a 'Green' cellphone. It is clearly noted that most consumers are willing to pay substantially high amounts to purchase 'green' cellphones. A large percentage, 20% consumers were willing to pay above RM500 once for the purchase of a 'green' cellphone. This was followed by 19% willing to pay RM 100-RM 200 and 17% willing to pay RM 70-RM100.

This may also be because the question specifically mentioned 'Pay Extra Once'. Hence, as it was not a periodic amount to pay, most consumers did not mind paying a large amount at the point of purchase.

In a study done by Milovantseva (Milovantseva, 2016) on a similar subject on a population from USA, the frequency of people willing to pay for green cellphones decreased as the amount willing to pay was increased. The minimum amount used was \$2.40 and the maximum was \$120. The study found that 22.3% respondents were willing to pay the minimum of \$2.40. A similar study was also done in Nigeria on consumer willingness to pay for green cellphones. The results showed that about 51% of respondents were willing to pay more than 20% of the premium price while approximately 27% were ailing to pay more than 10% but less than 20% of the premium price (Nnorom, Ohakwe, & Osibanjo, 2009). This is quite contradictory with a study done among Californian households who were willing to pay no more than 1% above the price of green computers or cellphones (Saphores et al., 2007).

In order to determine why consumers did not want to purchase a 'green cellphone', three options were provided to the survey takers. 82.6% answered that they do have some interest in 'green' cellphones but would not pay anything extra to buy

one. 18.9% stated that they do not have any interest in ‘green’ cellphones and 8.9% stated ‘Others’ but did not chose to elaborate.

From these polls, we can conclude that most consumers’ are willing to pay for green cellphones. Although the amount they are willing to pay may vary, in general, the willingness to pay exists. From the 82.6% who stated that they are not willing to pay anything extra, the factor of price may play a deciding role.

### 6.3 Model Estimation using Binomial Logistic Regression

The result interpreted from the tables would be used to formally test the hypothesized relationships between the independent variables and the dependent variable. The ‘Willingness to Pay (WTP)’ which is the dependent variable is denoted as 1 which would then make ‘Not Willing to Pay (NWTP)’ to be denoted as 0. If the probability of WTP is 0.5 or more, the Null model predicts WTP. The estimation of WTP is as summarized in Table 6.10. The results show estimation for consumers’ who have a higher willingness to pay for green products. Indirectly, this can be inferred as determinants of willingness to pay for green products.

$$\text{The null Model is : } \ln\left(\frac{p}{1-p}\right) = \beta_o = 0.221$$

$$p = \text{probability of Not Willing to Pay} = \frac{\exp(0.221)}{1 + \exp(0.221)} = 0.445$$

The goodness of fit of the model is satisfactory, as depicted by the various diagnostic statistics such as Cox and Snell and Nagelkerke and Hosmer & Lemeshow test. Cox and Snell and Nagelkerke values are both defined as Pseudo R<sup>2</sup>. They represent an analogy to the R<sup>2</sup> value in the Standard Regression. For our study, we can



conclude that between 27% to 36% of the variation for willingness to pay can be explained by the model. The -2LL for this model (416.87) is what was compared to the -2LL of the previous Null Model in the Omnibus Test of Model Coefficients, which was also evidence that the new model was significantly better fit than the null model.

The Hosmer & Lemeshow test of goodness of fit still uses  $\chi^2$  but in this case the  $\chi^2$  we are looking for is a non-significant value. The tests reveal that the significance for this test for our study is 0.425 which is a non-significant and consistent with our model fitting the data. This Hosmer and Lemeshow test is not highly recommended but it can be used as a screener when trying to show overall fit of model.

As the Logistic Regression Model predicts correct classification, the statistical analysis shows that the prediction rate has increased from 55.5% in the Null Model to the now 72%, which is a marked improvement.

**Table 6.2: Estimation Results**

		B	S.E.	Wald	df	Sig.	Exp(B)	95% C.I. for EXP(B)	
								Lower	Upper
	Gender(1)	-.410	.284	2.076	1	.150	.664	.380	1.159
	Current_Occupation			13.943	2	.001			
	Current_Occupation(1)	-.401	.322	1.554	1	.213	.670	.356	1.258
	Current_Occupation(2)	-.1381	.372	13.810	1	.000	.251	.121	.521
	Ethnicity			18.577	4	.001			
	Ethnicity(1)	1.460	.435	11.252	1	.001	4.306	1.835	10.107
	Ethnicity(2)	1.083	.305	12.621	1	.000	2.953	1.625	5.368
	Ethnicity(3)	1.446	1.329	1.184	1	.277	4.246	.314	57.412
	Ethnicity	-.100	.609	.027	1	.869	.904	.274	2.983
	Environment	.058	.068	.713	1	.399	1.060	.926	1.212
	Health	.125	.062	4.056	1	.044	1.133	1.003	1.279
	PeerPressure	.129	.039	11.101	1	.001	1.137	1.054	1.227
	Availability	.052	.085	.383	1	.536	1.054	.893	1.244
	Price	.021	.088	.057	1	.811	1.021	.859	1.215
	Knowledge	.114	.068	2.813	1	.093	1.121	.981	1.280
	Income			2.015	2	.365			
	Income(1)	.630	.483	1.703	1	.192	1.878	.729	4.839
	Income(2)	.139	.410	.116	1	.734	1.149	.515	2.565
	Edu_level			1.891	2	.389			
	Edu_level(1)	.786	.739	1.131	1	.288	2.195	.515	9.345
	Edu_level(2)	.945	.691	1.869	1	.172	2.573	.664	9.977
	AgeGroup			8.139	3	.043			
	AgeGroup(1)	-.1361	.492	7.662	1	.006	.256	.098	.672
	AgeGroup(2)	-.1432	.582	6.054	1	.014	.239	.076	.747
	AgeGroup(3)	-.1671	.704	5.639	1	.018	.188	.047	.747
	MaritalGroup(1)	.363	.340	1.142	1	.285	1.438	.739	2.800
	Constant	-.7758	1.571	24.373	1	.000	.000		
<p>Goodness-of-fit statistics:</p> <p>- 2 Log Likelihood                      416.874</p> <p>Model <math>\chi^2</math> ( df = 22)                      120.428</p> <p>Significance                                      p &lt; 0.01</p> <p>Cox &amp; Snell R Square                      0.265</p> <p>Nagelkerke R Square                      0.355</p> <p>Hosmer and Lemeshow Test              0.425 (significance)</p>									

Generally, with reference to Table 6.2, the variables that were significant were peer pressure, health and safety, occupation, ethnicity and age. The non-significant variables were availability, environmental awareness, the price of product, the knowledge available on the product, household income, education level and marital status. Several of the non-significant variables were contrary to expectations such as household income and price of product and even environmental awareness. These will be discussed in the following sections. The discussion on the socio-demographic variables will precede the discussion on the exploratory variables.

#### **6.4 Socio-Demographic Variables**

Based on the data in Table 6.2, the logit model seems to fit the data quite well. The variable current occupation, ethnicity and age group show significant overall effect. On the grouped variables, the reference for occupation is government job, and it is noted that those with private company jobs show a statistically significant contribution to willingness to pay (WTP) for green. However, when compared to those holding government jobs, the willingness of privately employed consumers to pay for green reduces 0.25 times. This is also seen where the  $\beta$  of privately employed consumers value is negatively correlated to willingness to pay. This may be because many government employees are exposed to the 'greening' of Malaysia through the works of several Malaysian ministries such as KeTTHA, MOSTI and even MITI. Hence, this exposure may encourage more government staff consumers' to be more informed and be more willing to go 'green' due to the benefits associated with it. Private employees may have price concerns and may not be as knowledgeable on green products as the government employees. Those who are self employed are also negatively correlated but do not show a significance.

A report in 1999 on consumers' in Hong Kong stated that the consumers claimed that they were influenced to go green by environmentally friendly groups and the government. This is evidence that the government is more likely to raise awareness on going green, and government employees being the first liners are more influenced (Chan, 2000). In Malaysia, we have the Green Government Procurement Practices that is being put to place. The Ministry of Finance spearheads this initiative. Although a policy on green procurement has not yet materialised, the Malaysian government is committed to give priority to green products and services when it comes to government procurement (Adham & Siwar, 2012; MOF, 2010). This also further exposes government employees to green purchasing.

Surprisingly, ethnicity seems to show significant contribution to WTP. With the Malay race as our reference, it is observed that both Indian and Chinese have higher probability to have willingness to pay for green products. As observed from the results, the Chinese are 4.31 (331%) times more likely to be willing to pay for green electronics and the Indians are 2.95 (195%) more likely to be willing to pay for green products. On the other hand, the Sabah Sarawak and other ethnicity groups did not reach the statistical significance of having the willingness to pay for green.

Age group shows a strong significance but all the age groups have negative B values, which means that they show less likeliness of willingness to pay when compared with the reference value between 18-24 years old. Those aged 25-40 were 0.26 times less likely to pay for green electronics, those aged 41-50 were 0.24 times less likely and those 51 years and above were 0.18 times less likely to pay for green electronics compared to their more younger counterparts. The conclusion from the age based results is that the older they become, the lesser the willingness to pay for a green cellphone.

This may be attributed to the fact that the older generation is less exposed to the information on green cells or even environmental consciousness or even maybe because for the older generation, a cellphone is just a means of communication, unlike the younger generation. For the younger generation, a cellphone is not just a means of communication. It has become a lifeline, as it is where they learn and socialize. Schools encourage the use of electronics in their curriculum.

On closer grounds, a study reported that Malaysian consumers' aged 20 and below were more inclined to purchase green products (Sinnappan & Rahman, 2011). With regards to ethnicity, the Chinese were found to be more willing to pay for green products (Ahmad et al., 2010; Goh & Wahid, 2014). Wahid et al reported in their study that the age group more inclined to purchase green is 21-40 year olds and of Chinese ethnicity (Wahid et al., 2011). This actually correlates with this study, which shows 21 – 40 year olds and Chinese ethnics as the most willing to pay for green cellphones.

Having said that, there have been studies, which have found inconclusive evidence with respect to influence of age in determining willingness to pay (Bollino, 2009; Do Paco et al., 2009; Kotchen & Moore, 2007; Yoo & Kwak, 2009)

Ethnicity was found to be inconsistent as a determinant where some researchers found that it was significant while others did not. In a study in 1991, the researchers did not find significant differences among ethnic groups across African American and Caucasians (Ellen et al., 1991). However, in 1997, a study reported that White consumers were more concerned with the environment, hence more willing to pay for green (Newell & Green, 1997).

In this study, the Chinese ethnics were found to be the highest willing to pay group followed by the Indians and Malay. All three major ethnic groups were found to be significant in willingness to pay but all three exhibited differences in their willingness. In a study done for Malaysians consumption of green food, Malay and Chinese consumers were found to be more willing to purchase green foods due to health reasons. However, their purchase decisions were affected by other factors such as household size, household income and location of stay as well (Quah & Tan, 2009). These results indicated that Malaysian consumers do value their health and other factors related with it. Their willingness to pay for green electronics may stem from the fact that they are conscious of the environment, which also directly contributes to health and well being.

Contrary to expectations, household income was an insignificant determinant in willingness to pay in our study. It was initially believed that household income would correlate with ability to pay which would be able to show us that a family with higher income would be able to pay more for green, hence they would be more willing to go green. However, this was not reflected in this study contrary to several other studies (Kotchen & Moore, 2007; Yoo & Kwak, 2009; Zorić & Hrovatin, 2012).

Income has been reported in many studies as a determinant for willingness to pay for green products. This is because income is believed to be the determinant of a person's budget limitations, hence the person's ability to pay. There are also studies, which have shown that statistically using income in a willingness to pay function depends also on the type of income used, whether relative income or absolute income who said that people's decisions depends not only on their income but their relative income which is their income in relation to others in the social community (Broberg,

2010, 2014). Relative income was first introduced by Duesenberry who said that people's decisions depends not only on their income but their relative income which is their income in relation to others in the social community (Duesenberry, 1949). This was also found true by a later study (Baumgärtner et al., 2016). The ability of a person to pay may depend on the absolute income but the willingness to pay depends on the relative income. Hence, even if a person has the ability to pay, it is not necessary that he or she is willing to pay (Magnani, 2000). This may be the case with the current study where the income is not statistically significant in determining WTP for green cellphones, hence cannot be used as a determinant.

Conclusively, as seen from this study, age and ethnicity does show a significant contribution in assessing consumers' willingness to pay for green electronics. In a 2016 study among American consumers', the researcher found that income, education, age and household size influenced the willingness to pay for green cellphones. However, it was the lower income household (\$50,000 and less per annum) that was more willing to pay compared to those with annual income between \$50,000 and \$100,000. On education, the consumers' with a higher level of education were found to be willing to pay for green cellphones compared to those with a Diploma (Milovantseva, 2016). In the present study, the income and education variable were not significant in determining willingness to pay. The result for age in our study showed that consumers' aged 18-24 were most likely willing to pay the highest compared to those above 25 years old. However one study has found that consumers' aged 45-59 were more inclined to pay for green cellphoned compared to other age groups (Milovantseva, 2016). Another study found that age was negatively related to willingness to pay for green electricity (Zorić & Hrovatin, 2012). The same study also found that gender did not play a significant role in determining willingness to pay, as did our study. Kaufmann reported that demographic

profiles such as age, ethnicity and income did have an impact on consumers' green purchasing behaviour (Kaufmann et al., 2012). Royne et al (2011) reported that younger consumers' were more willing to pay for green products compared to their older counterparts. The researchers explained this in terms of the educational information on environment and green purchasing that are disseminated better among younger individuals compared to older ones.

## **6.5 Exploratory Variables**

There were six exploratory variables in our study. All the variables were selected based on past research and results (Mohd Suki, 2015, 2016; Sriwaranun, Gan, Lee, & Cohen, 2015; Suki, 2013). The variables were not necessarily confined to studies in Malaysia. From the six variables we analysed, only two variables were statistically significant in determining willingness to pay among Malaysians. Peer pressure and health and safety were found to be significant determinants. Factors such as availability, environmental awareness, price of product and knowledge of product were not significant determinants according to our study.

### **6.5.1 Health and Safety**

Health was one of the variables that were significant as a determinant for willingness to pay for green cellphones in this study. In the survey questions, under Health and Safety variable, there were two questions, which were directly related to an individuals' health and its relationship to the environment. In a study by Royne, health was one of the three concerns, which were rated highly by the 91.2% of consumers', who were willing to pay more for eco-friendly products (Royne et al., 2011). In a study involving Malaysians, on the willingness to pay for green food, the maximum amount that respondents were willing to pay was 90% of the premium price and the least they



were willing to pay was 5% of the premium. The reason given for the willingness to purchase green was their concern about their health (Rezai et al.,2013). The high amount that consumers' were willing to pay is an indication that Malaysians are aware of the effects of non-green products towards their safety. They have the perception that buying green would mean their health fares better and at the same time the environment is taken care of.

An earlier study reported that researchers might have misunderstood that the intention for consumers' willingness to purchase green products may have been due to environmental concerns. However, it may also have been due to the fact that green foods, especially, have positive effects on an individual's health (Peattie, 1999). We believe that both may be true, that consumers' now are willing to pay for green due to health concerns and environmental concerns. If a consumer is aware of the detrimental effects non-green products have on their health, they may also be aware of its effect on the environment. Consumers' are trying to become more environmentally friendly, and lead a more sustainable life as they are also concerned about their health in the long run.

A study by Sinnappan (Sinnappan & Rahman, 2011) reports that Malaysian consumers' regard their health very highly and would take serious measures to ensure their health is well taken care of at all times. Hence, if consumers' were informed of the seriousness of the country's environmental problem, and how it related to their health in the long term, the chances of the consumers being environmentally aware and willing to pay for green may increase. This result, however, contradicts the finding by Lee (Lee, 2008) who found in a Hong Kong study that highlighting environmental problems did not increase environmental awareness but created ignorance among the young people.

Generally, health concerns are related to physical surroundings as well. People want their environment to be clean, with good air and water quality and a healthy quality of life. A healthy place of living will definitely promote healthy communities, which in turn would ensure clean and quality living. People who value their health, with the correct education and knowledge dissemination, can also learn to see the relationship between health and environment. A healthy environment would also mean a healthy lifestyle. Creating a healthy lifestyle is not just through green food consumption, but also through creating a clean environment – free of harmful chemicals.

Personal risk is also associated with health and safety. The perception that personal risk may increase the behavior of consumers' to be more willing to pay for green was studied by Martin (Martin & Simintiras, 1995), who reported that safety and health concerns are indeed the strongest factors in predicting consumers' attitude towards sustaining the environment. Another Malaysian study also corroborates with this finding. This study reported that Malaysian households hold safety and health concerns as the key in choosing a green electronic for the household (Hassan & Nor, 2013). This study was conducted among Malaysian who live in a medium to high-class area and who are most probably aware of the negative implications of lead and toxic materials and electronics. Although safety is highly regarded, focus should also be given to the information dissemination about the products to the target group. If the consumers' had no knowledge on the toxic and lead infested materials used in electronics, the chances of them being aware of the implications on their health may be very low.

Most consumers are aware of organic food. They understand that organic means chemical-free which is good for their health as there would be no build up of toxins in the body. Hence, they choose to purchase green food. Likewise, consumers' who are aware of the negative affects of chemicals and heavy metals in electronics would also be aware of its implications to health and would prefer to purchase green electronics. Several studies were also found to report this perception (Afroz et al., 2013; Huang et al., 2006; Liu et al., 2009).

### **6.5.2 Environmental Concern**

The non-significant results obtained on environmental awareness in our study were quite surprising, as many studies have reported a positive relationship between environmental concerns/awareness and willing to pay for green. However, there were studies that proved otherwise that there is no relationship between environmentally responsible behavior and values about environmental issues (Pickett-Baker & Ozaki, 2008).

Based on the survey questions for environmental concerns and awareness, individuals like to engage in activities, which would portray their actual identities. Mannetti et al (Mannetti et al., 2004) reported in their study that an individual who portrays environmentally responsible behavior has the intention to engage in green purchasing. This environmentally sensitive behavior has also been found as a determinant for green purchasing among young people in several other studies (Cheah, 2009; Lee, 2009).

Generally, consumers who have concern for the environment would act in ways that show their concern for the environment. They would believe that their actions

would somehow improve the environment and they would try to be as environmentally friendly as possible by purchasing green products or be willing to pay for green products. Consumers' may also feel that they are morally obligated to protect the environment. Stern has reported that moral obligation does reflect pro-environmental behavior (T. B. Chen & Chai, 2010; Stern, 2000).

A Kuwait study found that environmental knowledge and concerns did have an impact on consumers intention to purchase green (Mostafa, 2009). Several studies also found that environmental concern is linked to willingness to pay for green products (Biswas et al., 2000; Cornelissen et al., 2008; Mainieri et al., 1997). Mainieri et al. (1997) insisted that when consumers are concerned about the environment, they would most definitely be willing to purchase green products . A later study in 2005 found that environmental concerns is directly proportional to willingness to purchase green products (Kim & Choi, 2005).

At the same time, there are studies reporting either a negative or a weak relationship between consumers' environmental awareness and willingness to purchase green (Axelrod & Lehman, 1993; Berger & Corbin, 1992). These studies reported that although people are concerned about environmental deterioration, it does not reflect that they are willing to purchase green products. This then is reflected in the inconclusive relationship between environmental concern and willingness to pay for green (Ali, Khan, Ahmed, & Shahzad, 2011; Dunlap, Van Liere, Mertig, & Jones, 2000; Kollmuss & Agyeman, 2002).

The insignificant statistical result of this variable in the present research falls into the inconclusive category. Majority of consumers may be willing to pay but it may

not be because of their knowledge on environmental awareness as it has been reported that it is not necessary for consumers' with environmental awareness to be willing to go green or not. It is just a general consensus.

### **6.5.3 Availability and knowledge of product**

In general, for a product to be in demand or for a product to be purchased by a consumer, the product needs to be readily available and there needs to be enough information on the product for a consumer to be able to make an informed purchase decision. The results for availability of product and available knowledge on product are not significant as a deciding factor to be willing to pay for green electronics. This may be because the respondents in this study felt that they already have enough information about the product that they would like to buy, in this case cellphones or they are just not interested to buy green cellphones. Another reason may be the lack of green cellphone availability in many locations.

At the same time, a study has reported that lack of availability does influence consumers' purchase decisions (Ismail & Panni, 2008). In addition, the researchers also found that availability of information on advertised green products also mitigates product purchase. In addition, the lack of information influences the consumers' pro-environment behavior as well. This means that if a consumer does not have the required knowledge about a green product or even about the negative effects of non-green products on the environment, he or she is not going to pay much attention to its effect to the environment. For the consumer, there is no difference whether he purchases green products or not. There has been empirical evidence proving that consumers' also face difficulty in obtaining these green products due to lack of information about them (Banyte et al., 2010; Brown & Wahlers, 1998; Pickett-Baker & Ozaki, 2008). Several

studies also report that the information available on the availability and advantages of the green products makes the consumer more pro-environment (Banyte et al., 2010; Pickett-Baker & Ozaki, 2008). This is a fact that cannot be denied as it is only through knowledge that one can make sound decisions. Mainieri reported that one of the reasons consumers are not environmentally conscious is because of the lack of information about the available green products (Mainieri et al., 1997).

#### **6.5.4 Price of Product**

In many instances, price of a product plays a significant role in decision making. Regardless of whether it is to purchase a notebook or a washing machine, price is a contributing factor. This is also what has been reported in several studies (Alford & Biswas, 2002; T. B. Chen & Chai, 2010; Mazumdar & Monroe, 1990). Hence, it would be expected that the price of a green product, in this case a cellphone, would significantly affect a consumers' willingness to pay. Any price conscious consumer would opt to pay for a fair price, better yet, the lowest price for a particular good. In addition, price conscious consumers were also found to be more knowledgeable about a product as they would read labels and do their search on a particular product before deciding to purchase (D'Souza et al., 2006). This is an important point as these consumers who are price conscious, and be more knowledgeable on green products and its advantages, not only to the environment but also to them, would be more inclined to buy a green cellphone as it is not just the price that would matter but the added features as well.

Having analysed the price conscious consumer, studies have also shown that consumers' who are environmentally concerned become less price conscious when it comes to green products (Sriwaranun et al., 2015; Vlosky et al., 1999). Price would

seem to matter to them but if they are informed of the value they obtain for the amount they pay, price would take second place.

It is very interesting to note the behavioural pattern in Malaysia. This is not surprising as Malaysia is the first country in the Asian region to pioneer environmentally responsible behaviour. This was pledged by the Malaysian Prime Minister to reduce carbon emissions by 40% by the year 2020 at the 15<sup>th</sup> Conference of Parties (COP 15) in Copenhagen in 2009. Hence, it would be wrong to say that Malaysians are unaware of the ‘green revolution’. Studies have shown that price did not seem to be a major concern for Malaysians to purchase green products (Goh & Wahid, 2014). There are reports stating that if the price of a green product decreases, the demand and willingness to pay for the green product would increase (Mohamed et al., 2014). This is to be expected as price is usually a concern when making purchases, what more when the price for environmental friendly product when it is sold at a lower price than expected.

Contrary to the found reports, the present research show that price of the product is insignificant in determining willingness to pay for green cellphone. These reports conform to the results by Goh & Wahid. In general, Malaysians are becoming more and more aware of sustainability and going green. This is well proven in our study.

### **6.5.5 Peer pressure**

In everyday language, peer pressure would refer to influence by a certain peer group. Most adolescent Malaysians are eager to ‘belong’ to certain groups, be it a social class or a team in sports. Hence, how others in the group perceive them becomes very important. This is also true for ladies groups or clubs. Social status is highly regarded and because of this, people tend to purchase things or dress to belong to these

socialite groups. Branding becomes important. However, how do we equate peer pressure and willingness to pay for green products?

Social influence also known as peer pressure is believed to affect mainly the younger generation who are easily influenced by their peers. These can be their role models or someone they look up to, such as friends, family members or experts in the field who have a certain amount of influence on the individual (Posri, 2014). This exertion of peer pressure can be defined as shared values or thoughts or beliefs (Chen - Yu & Seock, 2002). At the same time, peer pressure can also create a feeling of guilt if the individual does not do as asked by the group or friend. Hence, the individual would then experience a behavioral shift. Peer pressure can also become psychological for some if they feel compelled to follow a certain group (Cohan, 2009). As much as they may not want to, they would do something just to feel accepted.

The statistical data shows a significant relationship between peer pressure and willingness to pay for green cellphones. This is in line with other studies on Malaysian consumers', which report that peer pressure is significant in determining consumer purchase decisions (Mei, Ling, & Piew, 2012; Wahid et al., 2011). In a study among Hong Kong adolescents, social influence was the highest determinant for green purchasing behavior (Lee, 2008). This was reiterated by Kalafatis who agreed that social influence was the highest determinant in willingness to pay for green products among UK consumers (Kalafatis et al., 1999). Peer pressure as a determinant has been found significant by any other researchers as well (Biswas et al., 2000; A. Chen & Peng, 2012; Suki, 2013; Teng, Wu, & Liu, 2015; Wahid et al., 2011).



With the number of reports stating the positive effects of peer pressure on willingness to pay for green, it was surprising to see that there were studies which contradicted the effects of peer pressure towards green purchasing (Schwepker Jr & Cornwell, 1991; Shamdasani et al., 1993; Sinnappan & Rahman, 2011; Wang, 2006).

Cordano et al. (2010) reported that the effect of peer pressure would depend on the sample frame used in the study. Studies using younger consumers' may find that social influence may be significant in determining willingness to pay but studies using older consumers' or adults may find that peer pressure is an insignificant determinant. This may be because adults are usually thought to make sane and informed decisions and not allow friends or family to influence them (Cordano et al., 2010; Sinnappan & Rahman, 2011).

The findings of our study can be explained by people's desire to feel included. The fear of exclusion allows them to conform to things they may not be willing to do but due to the peer pressure, are forced to do. Consumers often seek approval for their purchases from others in order to fit in to a social group. This happens when they want to say a statement about themselves. It has been said that what you consume says a lot about you, this is one such evidence. A Malaysian study of 239 international students has also reported a correlation between consumer consumption and material purchases (Heaney et al., 2005).

Goldsmith has reported that consumers' do feel a certain level of pressure to follow, depending on their own self-susceptibility to concede to social expectations just to belong to certain groups (Goldsmith & Clark, 2012; Goldsmith et al., 2010). This basically says that the more easily influenced consumers are, the more they will fall prey to peer pressure.

## **6.6 Summary**

The survey conducted to investigate consumers' willingness to pay for green electronics yielded 391 respondents. From the 391 respondents, 90 % were willing to pay for green cellphones. Most respondents who were not willing to pay for green cellphones were unwilling to invest in a green cellphone due to the price factor.

The analysis revealed that the significant determinants of willingness to pay for green cellphone was peer pressure, health and safety, occupation, ethnicity and age. With regards to age, consumers' most willing to pay in terms of age were those between ages 18 – 24. The prevalence of health and safety as a determining factor shows that Malaysians value their health and realize the increasing awareness on the relationship between environment and health.

## CHAPTER 7: CONCLUSIONS AND FURTHER WORK

### 7.1 Conclusions

Green energy and electronics have been in the limelight since the green revolution. The commitment of the Malaysian government at the Conference of the Parties (COP 15) in Copenhagen in 2009 showed the seriousness of the problem being faced today and made many Malaysians aware of green products and services.

In view of that, this two-pronged study reported in this thesis was to experimentally study the performance of biodegradable and non-biodegradable based polymer electrolytes in electrochemical devices and to discover through a survey, the level of awareness and determinants of Malaysian consumers' willingness to pay for green electronics.

This first part of the study aims to study and compare the nature and performance of biodegradable and non-biodegradable materials, which are then fabricated to become electrochemical devices. These devices are also subjected to characterization to compare their performance. For this study, poly (vinyl alcohol) (PVA) based polymer electrolytes were classified as the biodegradable entity and poly (ethyl methacrylate) based polymer electrolyte was designated the non-biodegradable one.

Ionic conductivity studies were performed on the biodegradable and non-biodegradable polymer electrolyte films to study which polymer electrolyte composition has the highest ionic conductivity. PVA-Mg(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub>-BmImBr polymer electrolyte system with 60 wt % of BmImBr portrayed the highest ionic conductivity of  $1.64 \times 10^{-3}$

$\text{Scm}^{-1}$ . We attribute this to the increase in the number of charge carriers and ion mobility in the polymer electrolyte matrix. In addition, the ionic liquid also acts as a plasticizer, which softens the polymer chain and increases its flexibility. The ion mobility in the flexible chain leads to an eventual increase in ionic conductivity. However, upon addition of 70 wt % of BmImBr, the ionic conductivity decreases to  $1.44 \times 10^{-3} \text{ Scm}^{-1}$ . This decrease is explained through the formation of ion pairs and aggregates that mitigate the ion movement in the conducting pathway making the polymer backbone less flexible. This reduces the ionic conductivity of the polymer electrolytes. The PEMA based polymer electrolyte system showed a systematic increase in the ionic conductivity upon addition of ionic liquid. The highest ionic conductivity obtained was  $1.80 \times 10^{-4} \text{ Scm}^{-1}$  upon addition of 40 wt % of BmImBr. The addition of 50 wt % BmImBr was detrimental since it was not possible to obtain a free-standing polymer electrolyte film. Results showed that biodegradable based polymer electrolytes exhibited higher ionic conductivity compared to non biodegradable based polymer electrolytes at room temperature.

The temperature dependent ionic conductivity studies were performed to study the mechanisms of ion transport in the polymer electrolyte matrix. For the PVA based polymer electrolyte systems, only the PVA 0 system was found to follow the Arrhenius theory. PVA 2, PVA 3, PVA 4, PVA 5 and PVA 6 was found to follow the Vogel-Tamman-Fulcher free volume theory that demonstrated the highest ionic conductivity at  $120 \text{ }^\circ\text{C}$  with 60 wt.% BmImBr and valued at  $(2.41 \pm 0.01) \times 10^{-3} \text{ Scm}^{-1}$ . Moving on to the PEMA based polymer electrolytes, it is noted that PEMA 0 obeys the Arrhenius theory. The PEMA systems incorporated with ionic liquid are found to be more inclined towards the VTF theory. VTF free volume theory combines the coupling effect of ionic hopping process with high segmental mobility in an amorphous phase. The temperature

dependent ionic conductivity also shows that the biodegradable polymer systems were better in performance compared to the PEMA based polymer electrolyte systems. To further support this point, the  $A_o$ , which is the pre-exponential constant that is proportional to the number of charge carriers, and B that is related to the rate the viscosity changes with temperature were examined. As expected, PVA 6, with the highest ionic conductivity had the highest number of charge carriers. In contrast, PEMA 4 was found to have lower  $A_o$ , value than PVA 6. A high number of charge carriers means that the diffusive ability of the ions is enhanced, which also contribute in increasing the ionic conductivity. This once again proves the superiority of the biodegradable polymer electrolyte in terms of number of charge carriers, which underwrites its higher ionic conductivity.

Thermal investigation, particularly DSC, was used to determine the Glass Transition Temperature ( $T_g$ ) of the sample, which is defined as the phase transition of the polymer from the hard, glassy state to an amorphous region, which is a flexible, soft and rubbery state. The elucidation of  $T_g$  is important for electrochemical device fabrication as it shows us whether a material is more amorphous or crystalline. PVA 6, the highest conducting polymer electrolyte was found to have  $T_g$  of  $-23.86\text{ }^\circ\text{C}$ . The  $T_g$  of PEMA 4, the highest conducting non-biodegradable system, was higher than that of PVA 6 with a value of  $-17.99\text{ }^\circ\text{C}$ . The importance of a low  $T_g$  is that the polymer backbone becomes more flexible due to plasticizing effect, which would favour a swift ion migration process, hence the higher ionic conductivity. It is noteworthy that the  $T_g$  value for the biodegradable polymer electrolyte is lower than the non-biodegradable polymer electrolyte showing that the biodegradable polymer electrolyte is more likely to be used in electrochemical devices due to its higher plasticizing ability, which would

give a higher ionic conductivity and may also improve the performance of an electrochemical device.

In this study, XRD was performed to investigate whether complexation took place in the biodegradable and non-biodegradable polymer electrolytes upon addition of  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  and BmImBr. For both PVA and PEMA based polymer electrolytes, upon addition of  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  into the polymer electrolyte – ionic liquid complex, the sharp crystalline peak of the salt was found to disappear. This proves the complexation of the polymer electrolyte-  $\text{Mg}(\text{CF}_3\text{SO}_3)_2$  and BmImBr. It also means that addition of the salt and ionic liquid has indeed increased the amorphous nature of the polymer electrolytes. In this investigation, both the biodegradable and non-biodegradable polymer electrolytes showed excellent complexation, proving its amorphous nature that enhances ionic conductivity of the polymer electrolyte.

Linear sweep voltammetry (LSV) was carried out to determine the maximum operational potential for a polymer electrolyte for energy storage in the EDLC, which will be fabricated. The maximum operational window for PVA 6, the highest conducting biodegradable polymer electrolyte was obtained as 2.6 V. On the other hand, the electrochemical potential window for PEMA 4 was obtained to be 1.3 V. An unexpected observation made was that the electrochemical potential window for PEMA 0 (without addition of ionic liquid) and PEMA 4 (with 40 wt% ionic liquid) was the same. From both the LSV results, we note that although the PEMA 4 showed the highest ionic conductivity, in terms of electrochemical potential window, the addition of ionic liquid did not cause an improvement.

A wider electrochemical potential window is important as it determines the “safe” maximum operating temperature of that particular electrochemical device and how long the device stays stable. Hence, the higher it is the more stable the device will be. For example, the electrochemical potential window of lithium ion batteries is 3 V, which is close to the window of our biodegradable polymer electrolyte. Once again, the experimental results have revealed the superiority of the biodegradable polymer electrolyte system. From both the LSV results, it was noted that although the PEMA 4 showed higher ionic conductivity compared to PEMA 0, in terms of electrochemical potential window, the addition of ionic liquid did not cause an improvement.

The next part of the study was the fabrication of electric double layer capacitors (EDLC). The biodegradable EDLC was fabricated using PVA 6 and the non-biodegradable device was fabricated using PEMA 4. Cyclic voltammetry (CV) was performed to evaluate the performance of the electrochemical capacitor in relation to its electrochemical behavior and charge storage. PVA 6 based EDLC showed a specific capacitance of  $45.95 \text{ Fg}^{-1}$  and PEMA 4 based EDLC displayed a capacitance value of  $7.34 \text{ Fg}^{-1}$ . As mentioned in the discussion, a rectangular shape for a CV voltammogram is deemed ideal and this was shown by the PVA 6 voltammogram. The voltammogram for PEMA based EDLC was leaf shaped indicating the low ionic conductivity and poor interfacial contact of the polymer electrolyte. It is important for a polymer electrolyte to have good interfacial contact especially in fabrication of devices. In EDLCs, the excellent interfacial contact between the polymer electrolyte and the electrode increases in the energy storage capability of the EDLC. This was clearly evidenced by the CV value obtained for the biodegradable polymer electrolyte, which was nearly 7 times higher than the value obtained for the non-biodegradable electrochemical device.

The electrochemical storage and pattern of EDLC can also be evaluated through constant current charge-discharge. In addition the stability can also be evaluated. For the biodegradable polymer electrolyte based EDLC, a specific capacitance of 45.25  $\text{Fg}^{-1}$ , energy density of 4.00  $\text{Wh kg}^{-1}$  and power density of 61.33  $\text{kW kg}^{-1}$  was obtained. As expected, these values are in line with the positive results obtained from CV. However, the attempt to carry out an evaluation of electrochemical storage capability of the fabricated EDLC using PEMA 4 did not yield any results. This was found to be interesting as the only possible reason was because the experiment revealed that there was poor interfacial contact between the electrode-electrolyte. The PEMA 4 sample was totally solid with no adhesive property whatsoever. This poor interfacial contact could not conduct a charge of the device, what more a discharge. It was also clearly seen in the specific capacitance analysis that the capacitance value obtained was low. It can be concluded that although a reasonable ionic conductivity was obtained for PEMA 4, its physical attributes prevented it from giving good performance when fabricated into a device.

The motive of this research was to compare the biodegradable material and non-biodegradable materials with regards to performance upon device fabrication. To prove that biodegradable materials are not only safe for the environment but also in terms of performance, an electric double layer capacitor was selected as the device to fabricate as EDLC. Based on the results, it was established experimentally that biodegradable polymer electrolytes are better materials for use in electrochemical devices. In hindsight, not only are these biodegradable materials gentle on the environment, but can also improve the performance of an electrochemical device.



A survey was conducted to investigate consumers' willingness to pay for green electronics and define the determinants of willingness to pay for green electronics. From the survey obtained from 391 respondents, it was discovered that 90 % were willing to pay for green cellphones. It was also found that from the 90%, 20% consumers were willing to pay above RM500 once for the purchase of a 'green' cellphone. This was followed by 19% willing to pay RM 100-RM 200 and 17% willing to pay RM 70-RM100. When consumers' who were not willing to pay for green cellphones were asked why they were unwilling to invest in a green cellphone, 82.6% answered that they do have some interest in 'green' cellphones but would not pay anything extra to buy one. This suggests that although the awareness about importance of going green exists, the price factor still prevails as a determinant factor for decision making.

From the analysis done, it was revealed that the significant determinants of willing to pay for green cellphone was peer pressure, health and safety, occupation, ethnicity and age. The non-significant determinants were availability, environmental awareness, price of product, knowledge available on the product, household income, education level and marital status. Contrary to expectations, household income, price of product and environmental awareness showed non-significance in this study.

It was also found that government employees were more inclined to purchase green devices compared to private or self-employed consumers'. This was because of the many green procurement awareness campaigns that is being conducted by the Government of Malaysia currently.

With regards to age, consumers' most willing to pay in terms of age were those between ages 18 – 24. This may be because most consumers' of this age group

practically live with their cellphones. From lecture notes to online business, everything is within the touch of a button. Hence, it may be important for them to have the best cellphone with all the effects necessary.

Health and safety was a deciding factor in purchasing green cellphones among Malaysians. This is obvious because many Malaysians are aware of the health implications due to chemicals in food, air quality, water quality and a clean environment. Consumers' who are aware of the negative effects of chemicals and heavy metals in electronics would also be aware of its implications to health and would prefer to purchase green electronics. People who value their health, with the correct education and knowledge dissemination, can also learn to see the relationship between health and environment.

Peer pressure was another significant determinant in consumers' willingness to pay for green electronics. In addition to the positive effect, peer pressure can also create a negative feeling of guilt if the individual does not do as asked by the group or friend. This feeling can become psychological hence forcing a habit just to be accepted.

The second part of this study proved that many consumers are willing to purchase green electronics. Peer pressure and health and safety seem to be major determinants for consumers' to decide to purchase green. Consumers' not willing to pay for green claimed that although they are aware of negative environmental effects of non-green electronics, they were not willing to pay extra for green cellphones.

## **7.2 Further Work**

For further work, it is proposed that a comparative study involving PVA polymer blends and PEMA polymer blends may be able to shed the light on its further usage in electrochemical devices. In addition to that, using different ionic liquids may also have different effect when blended with these biodegradable and non-biodegradable systems.

On the survey part, the present study focused on working Malaysians' whereas many studies conducted among Malaysians is on student perception. It would be interesting to investigate the difference if a survey was conducted comparing the Gen Y, the age cohort born between 1984 and 1996, and Gen Z which comprises those born after 1997, in terms of their green purchasing habits and preferences.

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## LIST OF PUBLICATIONS AND PRESENTATION

1. **K.A. Francis**, Chiam-Wen Liew, S. Ramesh, K. Ramesh & S. Ramesh (2016) Ionic Liquid Enhanced Magnesium based polymer electrolytes for Electrical Double Layer Capacitors, *Ionics*, 22:919–925
2. **Katherine A. Francis**, Chiam-Wen Liew, S. Ramesh, K. Ramesh and S. Ramesh (2016). Investigation on the effect of Ionic liquid 1-butyl-3-methylimidazolium bromide on ionic conductivity of poly (ethyl methacrylate) based polymer electrolytes, *Materials Express*, 6: 252-258

### Presented in International Conference

1. **K.A. Francis**, Chiam–Wen Liew, S. Ramesh, K. Ramesh, S.Ramesh (2017). Ionic Liquid Enhanced Polymer Electrolytes for Environmental Friendly Electric Double Layer Capacitors. Paper presented at 232<sup>nd</sup> ECS Conference at the Gaylord National Resort and Convention Center, USA