

**DESIGN AND DEVELOPMENT OF THE ALUMINIUM-AIR BATTERY**

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**A project report submitted in partial fulfilment of the  
requirements for the award of Master of Engineering**

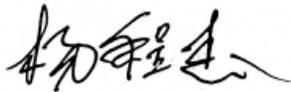
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**April 2021**

## DECLARATION

I hereby declare that this project report is based on my original work except for citations and quotations which have been duly acknowledged. I also declare that it has not been previously and concurrently submitted for any other degree or award at UTAR or other institutions.

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APPROVAL FOR SUBMISSION

I certify that this project report entitled “**DESIGN AND DEVELOPMENT OF THE ALUMINIUM-AIR BATTERY**” was prepared by **YONG CHENG JIE** has met the required standard for submission in partial fulfilment of the requirements for the award of Master of Engineering at Universiti Tunku Abdul Rahman.

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## **Acknowledgement**

I would like to thank my family who help me go through this part time master program, special thanks to my wife for taking care the kids during the weekend so that I can have study time and my employer to allow me to space time between work hours to attend the university.

I would like to thanks Dr. Bernard Saw for supervising me thought this project. Mr. Tan WC, Mr. Ho and Mr. Izhar for assisting me through the fabrication and testing of the battery.

## **Abstract**

Lithium-ion is have problem with its raw material availability and recyclability, lithium ion battery also have high cost and have safety concern. Although all this con for Lithium battery, it is still the dominant energy-storage technology for many application. Iron, zinc, magnesium, and aluminium, are some of the best material to be incorporate into the future batteries because of their abundance, capability of recyclability, low weight, cheap cost, and multiple electron redox properties. All this metal can be used as metal negative electrodes for metal–air batteries. Among the metal-air battery, the aluminium-air (Al-air) battery is candidate of next generation battery, especially in the field of electrical vehicles due to the high theoretical energy density (8100 Wh kg<sup>-1</sup>) Al-air batter have, which is much higher than the best performing lithium-ion batteries (LIBs).

In this project, an Al-air battery is being design and developed which consist of aluminium anode, air cathode, and electrolytes with casing. The material used for each component is being researched, compared and selected. The different manufacturing process of the battery is also being compared and the most viable method is used to build the battery. The cost of the material and manufacturing is also being look in to and the most cost efficient way to fabricate the unit was being used. Polarization test and discharge test were conducted using a VersaSTAT 4, a device which consist of the function of Potentiostat and Galvanostat to analyses the performance of the battery build.

The battery build have an open circuit voltage of 1.2V per cell, higher voltage is achieve by connecting the cell together, a three cell battery is build and have an open circuit voltage of 3.5V. The single cell battery can give off a maximum power of 0.015W and have a 175mAh capacity.

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# CHAPTER 1

## 1 Introduction

### 1.1 General introduction

Lithium-ion is have problem with its raw material availability and recyclability, lithium ion battery also have high cost and have safety concern. Although all this con for Lithium battery, it is still the dominant energy-storage technology for many application. Iron, zinc, magnesium, and aluminium, are some of the best material to be incorporate into the future batteries because of their abundance, capability of recyclability, low weight, cheap cost, and multiple electron redox properties. All this metal can be used as metal negative electrodes for metal–air batteries. (H. Schwarz,2004)

The energy density of Al-air is 8.1 kWh kg<sup>-1</sup> the theoretical voltage is (2.7 V) which is far higher than the best performing Lithium Ion Batteries currently available. Furthermore, aluminium is a very cheap and abundant material. Aluminium is also environmental friendly and have a high recyclability. (M. Nestoridi, et al. 2008). A critical weakness of metal-air to achieve commercial success is the severe self-discharge problem. In spite of huge breakthrough in corrosion inhibition, most metal-air batteries will self-discharge all its charge in the span of a month depending of the metal used as metal electrode in the metal air battery. On the other hand Li-ion batteries can maintain 95% of the battery charge after a month of storage. Until now most corrosion prevention methods either have insignificant effect on the suppression of the parasitic reaction unless the method come with the sacrifice of the power and energy density of the metal air battery. (Hopkins,et al, 2018.) Base on the current research and basic principle of aluminium air battery, an aluminium air battery is been design and using polarization test and constant current discharge test, the performance of the battery is being analyse.

## **1.2 Problem statement**

While many studies have been done on the aluminium air batteries, the application of the aluminium air batteries is still not yet common. A commercial viable aluminium-air battery is needed. To achieve a commercial viable aluminium air battery, this project an aluminium air battery is being design and develop. The project study available material and the manufacturing method and combining the two, the battery is being build. The cost of the material and manufacturing is also being look in to and the most cost efficient way to fabricate the unit was being used.

Polarization test and discharge test were conducted to analyses the performance of the battery build.

## **1.3 Aim and objective**

1. To design a multi-cell aluminium-air battery
2. To develop and fabricate a multi-cell aluminium-air battery
3. To evaluate the performance of aluminium air battery

## CHAPTER 2

### 2 Literature review

#### 2.1 Introduction

The increase in energy demands from modern lifestyle and industrial needs have been a huge driving force to keep progress in advanced energy research for many years. Tremendous effort has been put into research and creating new energy sources as well as energy storages with high capacity and compact in size so that it is portable, the energy source also need to be low cost and green. (Wang H et. al 2009)

Metal–air cells, featuring high energy density, have received many interest from the research community and have been predicted to be the next energy sources for electric vehicles, suitable for energy storage and can also be used as emergency power supply(Armand, M et al. 2008) (Winter M et al 2004)

Energy storage and conversion system with high energy density is develop to fulfill the demand and the need appliance that require portable energy. Metal such as iron, zinc, magnesium, and aluminium, are some of the best material for use in next-generation sustainable batteries. The reason is due to their abundance, the metals recyclability, the low weight of the metal, and cost of these metal., metal-air batteries is one of the most talk about energy storage system among all the new energy storage systems because of their high energy density and capacity. (M.A. Rahman et al. 2013) (D. Wang et al. 2015)

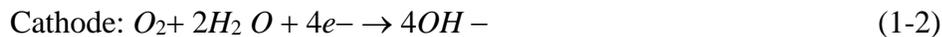
Table 2-1 Parameters of different metal-air batteries. (Y. Liu, et al, 2017)

Batteries	Theoretical Voltage (V)	Theoretical specific capacity (Ah Kg <sup>-1</sup> )	Theoretical energy density (kWh kg <sup>-1</sup> )	Practical operating voltage (V)
Li-air	3.4	1170	13	2.4
Zn-air	1.6	658	1.3	1-1.2
Mg-air	3.1	920	6.8	1.2-1.4
Na-air	2.3	687	1.6	2.3
Al-air	2.7	1030	8.1	1.2-1.6

Among the metal-air batteries, Al-air batteries hold highest potential for future large-scale energy applications because of the abundant of aluminium, to battery low cost and the high theoretical specific capacity of 1030Ah kg<sup>-1</sup>, which is the only lower than lithium-air battery and higher than magnesium base and zinc base metal air battery (Li Q et al. 2002) (H. Schwarz 2004) (M. Nestoridi et al. 2008)

The Al-air battery can produce sufficient energy and power for EV to handle long range journey and can generate acceleration in electrical vehicle comparable to gasoline powered cars. (H. Knickle 2002) H.Knickle also demostare that the aluminium anode only cost USD 1.1/kg if the reaction product is recycled. He also show that Al/air electric vehicles (EVs) can have higher total fuel efficiency than internal combustion engine vehicles (ICEs). Life-cycle of three different EV was also being compared by H. Knickle, the three candidate are Al/air EV, lead/acid EV and nickel metal hydride (NiMH) EVs. Among this three type of electrical vehicle only the Al/air EV projected to have long travel range which is similar to traditional petrol fuel car. (H. Knickle 2002). The aluminium-air battery has good discharge performance, the battery can provide stable power supply, and are able to meet the demand of power (Zu Liu et al 2016). In 2014, Phinergy Company are able to produce an electric vehicle based on Al–air cells that have a travel range of 1100-mile. ( Edelstein, S., 2014)

Fig. 1 is an illustrates basic diagram of a simple Al-air battery, The Al-air battery is composed of an aluminium/metal anode, an air cathode, and a electrolyte, a typical electrolyte is a aqueous alkaline solution such as sodium hydroxide (NaOH) or potassium hydroxide (KOH) or sometimes sodium chloride (NaCl) solutions can be used (N. Tanaka 2011) .The electrochemical reactions at the electrodes are listed below (M. Mokhtar et al. 2015):



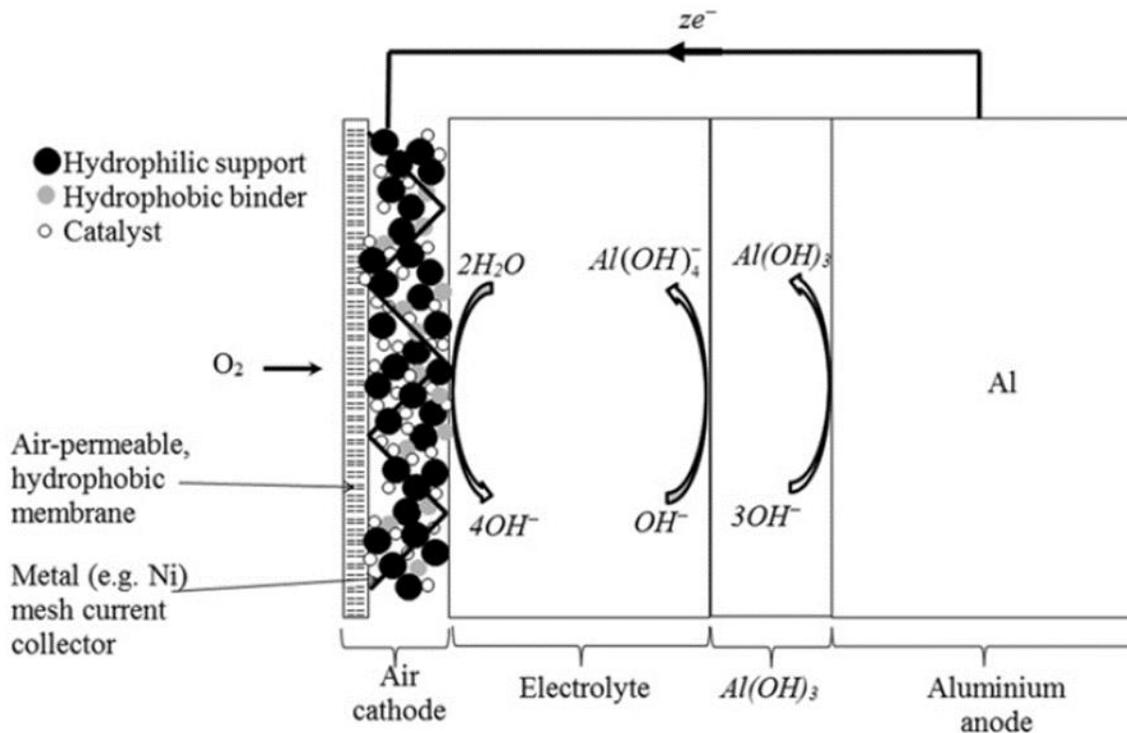


Figure 2-1 Illustration of the structure of an aluminium air battery using a 3-layer gas diffusion electrode and, in this case, pure aluminium as anode with its passive hydroxide layer. (D.R. Egan et al. 2013)

## 2.2 Anode materials

The electrochemical properties of pure aluminium make it a prime candidate as an anodic material for metal-air battery. Thermodynamically the pure aluminium anode have a potential of -1.66 V in neutral saline solution and -2.35 V in aqueous solution. But, the actual open-circuit potential is noticeably higher, this is cause by the four factor below (A.A. Mohamad 2008) (J. Bernard et al. 2006) :

- (i) The dissolution and formation of an initial Al<sub>2</sub>O<sub>3</sub> then the formation of Al(OH)<sub>3</sub> film,
- (ii) The production of Al<sup>3+</sup> species,
- (iii) The evolution of Al(OH)<sub>4</sub> and Al(OH)<sub>3</sub>
- (iv) parasitic corrosion which form hydrogen



(M. L. Doche et al. 1999) .

The aluminium surface is corroded by water and forms a passivation layer, this progresses to the breakdown of Al-air batteries. The protective oxide film causes failure to Al-air batteries. Oxide film forms on aluminium surface when aluminium reacts to air or aqueous solutions. The oxide film will slow down aluminium activation. (L. Fan et al. 2015) (M. Pino et al. 2016). The self-corrosion has a negative impact on the capacity and discharge efficiency of the battery. The passivation layer can be etched off by strong alkaline solutions, and aluminium anode can have good discharge performance in alkaline solution particularly it will have better performance at high discharge rates. But, alkaline solutions cause aluminium to undergo a high amount of self-discharge reactions with the generation of large quantities of hydrogen gases. (A. Elango et al. 2009)

### **2.2.1 Pure Aluminium**

(Young et al., 2015), study that when 99.5% purity aluminium is used for the Al-Air batteries have lower battery performance compared to when 99.99% pure grade aluminium. This is caused by impurities of the aluminium and formed impurity complex layer that contain Fe, Si, Cu and other material. These complex layers decline the battery voltage on open circuit status, the impurity also has an impact on discharge current and battery efficiency at 1.0 V. This can be overcome by reducing discharge voltage to 0.8 V which will dissolve the impurity complex layer in the 99.5% (2N5) grade aluminium anode, which can improve the efficiency of the battery of the lower grade aluminium-air batteries by 57%. The cheaper 2N5 grade Al is a viable anode because it is much cheaper than the 4N grade 99.99% aluminium and with some modification to the discharge voltage the overall efficiency of the 2N5 Al-air battery can be comparable to the 4N grade Al-air battery.

### **2.2.2 Aluminium alloy**

The parasitic corrosion, or self-discharge, has a negative impact on the anode efficiency and needs to be reduced or suppressed to keep the capacity loss at a minimum. By doping different metals in Al, such as Mg, Zn, Pb, Ga, In, Mn, Sn, Hg and Tl, the parasitic corrosion can be suppressed, this method is considered as the most popular and effective one. The parasitic corrosion is suppressed by the doped elements which have high hydrogen evolution overpotential, lower melting temperatures and higher solubility degree in Al. (S. Z. El Abedin et al. 2004) (M. Nestoridi et al. 2009) (Smoljko et al. 2012) (D. R. Egan et al. 2013)

Activation of aluminium can also be attained by adding small amount of suitable metal cations, sample of the metal cations are  $\text{In}^{3+}$ ,  $\text{Ga}^{3+}$ ,  $\text{Hg}^{2+}$  or  $\text{Sn}^{4+}$ , to the electrolyte. The electrochemical behaviors of Al (99.999%), Al (99.61%), Al-Sn, Al-Zn and Al-Zn-Sn alloys have been analyzed in 0.6 M NaCl solutions with  $\text{Sn}^{2+}$  ions added. (Shayeb et al 2001), found that the addition of  $\text{Sn}^{2+}$  causes activation of Al-Zn and Al-Zn-Sn alloys differ depending on  $\text{Sn}^{2+}$  concentration added.

(Jingling Ma et al 2014), build a metal-air battery based on Al, Zn, Al-0.5 Mg-0.02 Ga-0.1 Sn and Al-0.5 Mg-0.02 Ga-0.1 Sn-0.5 Mn (wt%), the battery performance is then being analyzed. Self-corrosion rate is used to study the corrosion behavior. The results show that Al-Mg-Ga-Sn-Mn is more active than Al, Zn and Al-Mg-Ga-Sn anodes. He found out that Al-Mg-Ga-Sn alloy perform better than Al-Mg-Ga-Sn-Mn while pure Al have the worse (fastest) self-corrosion rate. It is also found that Al-Mg-Ga-Sn-Mn can achieve highest operating voltage and better anodic utilization compared to other candidate he choose.

Bi, Pb and Ga are doping elements that have beneficial effect for Al electrode. Firstly Bi can precipitate along the grain boundaries within aluminium crystalline structure. This cause the activation of the Al anode in alkaline electrolyte (H. Moghanni-Bavil-Olyaei et al. 2016). Pb can slow down the hydrogen evolution effectively in Al anode. Pb can also speed up the sapling of discharge product of aluminium, thus enhance the discharge activity of Aluminium. (N. G. Wang et al. 2010) Ga can cause thinning of the passivized oxide film and activate the surface of aluminium. (M. Srinivas et al. 2016)

(Qin Wang et al 2017), develop an Al alloy base on Al-Bi-Pb-Ga. He is able to achieve higher electrochemical activity and better power density compare to pure aluminium in both neutral salt and alkaline electrolytes. Superior performance of Al-Bi-Pb-Ga is cause by chemical composition as well as the microstructure feature of the alloy.

The inhibition of parasitic-corrosion in KOH solution is linked to deposition of hydrogen evolution overpotential elements such as Bismuth, Lead and Gallium deposit on the surface of aluminium. This accelerated the dissolution of the Al matrix *via* micro-galvanic effect is cause by the segregate phases formed by Bismuth, Lead and Gallium. Al-Bi-Pb-Ga alloy is good potential anode material for Al-air battery due to the strong electrochemical activity as well as high anodic efficiency in saline electrolyte and alkaline electrolytes. (Qin Wang et al 2017)

### 2.3 Air cathode

The air cathode is one of the most costly component of metal air batteries and is one of the key factor to determine the cell performance. (V. Neburchilov et al. 2010) (H. Moghanni-Bavil-Olyaei et al. 2016). The air cathode is have three typical component.

1. Carbon material and a hydrophobic binder is used to construct gas diffusion layer. This allow permeable for air and do not allow permeable of water.
2. The current conductor is made using a good conductor such as Ni or Cu, and connect to the external circuit.
3. The catalytic active layer is made from electro catalyst, carbon material and binder. This layer is use to maximize the oxygen reduction reaction (ORR).

Typical air cathode/ gas diffusion electrode have multi layered structure of the gas diffusion layer and active layers complete with conductive mesh as charge collectors. (Z.Q. Fang 2006)

Aluminium air batteries is able to achieve high current densities, make it a good candidate for application that require high power density with short duration of discharge time. The consistency of binder and carbon is control across the air cathode, this form a bimodal pore network which optimize the oxygen diffusion and electron transfer for the electrode (S.K. Tiwari et al. 1995) (H. Arai, et al, 2000) (R.N. Singh et al. 2002). Sintering times along with lamination pressure used during manufacture is been study by (J. Prakash et al. 1990), to maximize the porosity of the air cathode.

The resistance correspond with peroxide diffusion from carbon to catalyst can be minimized by utilizing the large surface area carbon (S. Muller et al. 1999) The high activity of perovskite electrodes is part cause by the encouragement of the direct four- electrode ORR. The high activity is also due to catalytically decomposing the peroxide generated around air cathode (Y.K. Shun et al. 2000).

The composition of the air cathodes catalyst are based on individual oxides or mixture of oxides, consist of spinal, perovskite or pyrochlore structure, the typical oxide used are  $\text{MnO}_2$ ,  $\text{Co}_3\text{O}_4$ ,  $\text{La}_2\text{O}_3$ ,  $\text{NiCo}_2\text{O}_4$ ,  $\text{LaMnO}_3$ , and also noble metal such as platinum and silver. (Vladimir Neburchilov, et al. 2010), the maximization of ORR activities and chemical stability are able to be achieve by these composition in an alkali electrolyte.

The design of the air cathode which include type of carbon, the binding agents, charge collectors, Teflon membranes, thermal treatment of the gas diffusion layers (GDL) and catalyst layer have a strong impact on the performance of the batteries. (J.J. Martin et al. 2009)

A high current density 3000mA cm<sup>-2</sup> were attainable with the application of pure oxygen instead of ambient air,(X.H. Yang. Et al. 2010) these high current density can also achieve via elevated pressure (Y. Kiros et al. 2004).

### 2.3.1 Oxygen reduction reaction

The cathodic reaction in the Al-air system is centralise around oxygen reduction reaction (ORR).

The dual pathways for the oxygen reduction in aqueous alkaline electrolyte are: a four electron pathway, as well as a peroxide two – electron pathway.

A direct four electron pathway (2-1) is the preferable route. The two - electron pathway is expressed by (2-2), (2-3) and (2-4). When reactions (2-2) and (2-3) are happening at a high rate, the ORR occurs directly through the four-electron transfer pathway. (E. Yeager, 1986) (F.Li et al. 2013).

Formula for the direct four electron pathway



In the direct four electron pathway oxygen directly reduce to OH-

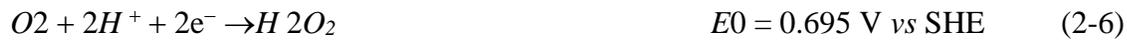
Formula peroxide two – electron pathway



While in the peroxide two electron pathway an initial reduction to HO<sub>2</sub><sup>-</sup> then is follow by the reduction from HO<sub>2</sub><sup>-</sup> to OH<sup>-</sup>.(E. Yeager, 1986)

The peroxide pathway of the ORR occur more frequently in alkaline solution and the direct four electron pathway of the ORR proceed by dissociative absorption of the metal catalyst surface (F.Li et al. 2013).

In acidic electrolyte, the two pathway which is four-electron (2-5) and two-electron (2-6, 2-7, and 2-8) is describe below :



## 2.4 Electrolyte

Aqueous electrolytes have categories corresponding to their pH. Alkaline categories range from ( $7 < \text{pH} \leq 14$ ), neutral salt electrolytes (NaCl solution) ( $\text{pH} = 7$ ), and acidic category have range of ( $1 \leq \text{pH} < 7$ ). The most popular electrolytes used for Al - air batteries is alkaline solutions. Sodium hydroxide or potassium hydroxide are some of the common alkaline electrolyte. (D.R.Egan et al. 2013) (Y.Liu et al. 2017)

### 2.4.1 Neutral salt electrolytes

Natural saline electrolyte is also been used as they have a lower open circuit corrosion rate and is safer to handle and have less health impact if been expose to human.

The potential values for pure Al electrodes is around 0.65 V–1.1 V when neutral salt solutions is used for the Al-air battery (S. Z. El Abedin et al. 2004). The redox potential values of Al will be depend on the concentration of the neutral salt solutions as well as the operating temperatures (S. Gudić et al. 2005). Binary and ternary alloys of Al are can be used to improve the performance of Al-air batteries that use neutral salt electrolytes (J. Ma et al. 2013) (L. Wang et al. 2013)

### **2.4.2 Acidic electrolytes ( $2 \leq \text{pH} < 7$ ).**

Sulphuric acid is also been used for aluminium air cells. Theoretical cell potential difference in an acidic electrolyte are higher compared to alkaline electrolytes and can vanquish the anodic dendrite formation and carbonate formation around air electrode . (M.Rota et al. 1995)

Aluminium is not stable in most acidic solutions, causing serious corrosion problems. (S. Miller et al. 1996) The application of acidic electrolytes in Al-air batteries is not recommended. (D.R. Egan et al. 2013)

### **2.4.3 Alkaline electrolytes ( $7 < \text{pH} \leq 13$ )**

Alkaline electrolyte have higher conductivity, it also have hi  $\text{Al(OH)}_3$  solubility compared to saline electrolyte, this allow Al-air battery to achieve higher cell power and higher energy density, making alkaline solution the best candidate among the acidic, neutral and alkaline electrolyte.

The common alkaline electrolyte is NaOH and KOH. Their peak electrolytic conductivities is  $4 \text{ mol dm}^{-3}$  for NaOH and  $7 \text{ mol dm}^{-3}$  for KOH. KOH shows superiority to NaOH solution for oxygen reduction due to the higher solubility for oxygen, higher oxygen diffusion coefficient and lower viscosity.  $7 \text{ mol dm}^{-3}$  KOH also have higher solubility limit of the reaction product aluminate compare to  $4 \text{ mol dm}^{-3}$  NaOH. (D.R. Egan et al. 2013)

However Al-air battery using KOH solution cannot recycle alumina through the Industries Hall-Heroult process. This will have an impact of the recyclability of the Al-air battery that uses KOH (Qi Li et al. 2002)

Impurity of electrolyte will generate preferential sites for hydrogen evolution at aluminium surface, initiating the corrosion and electrode potential. ( M.L. Doche et al. 1999)

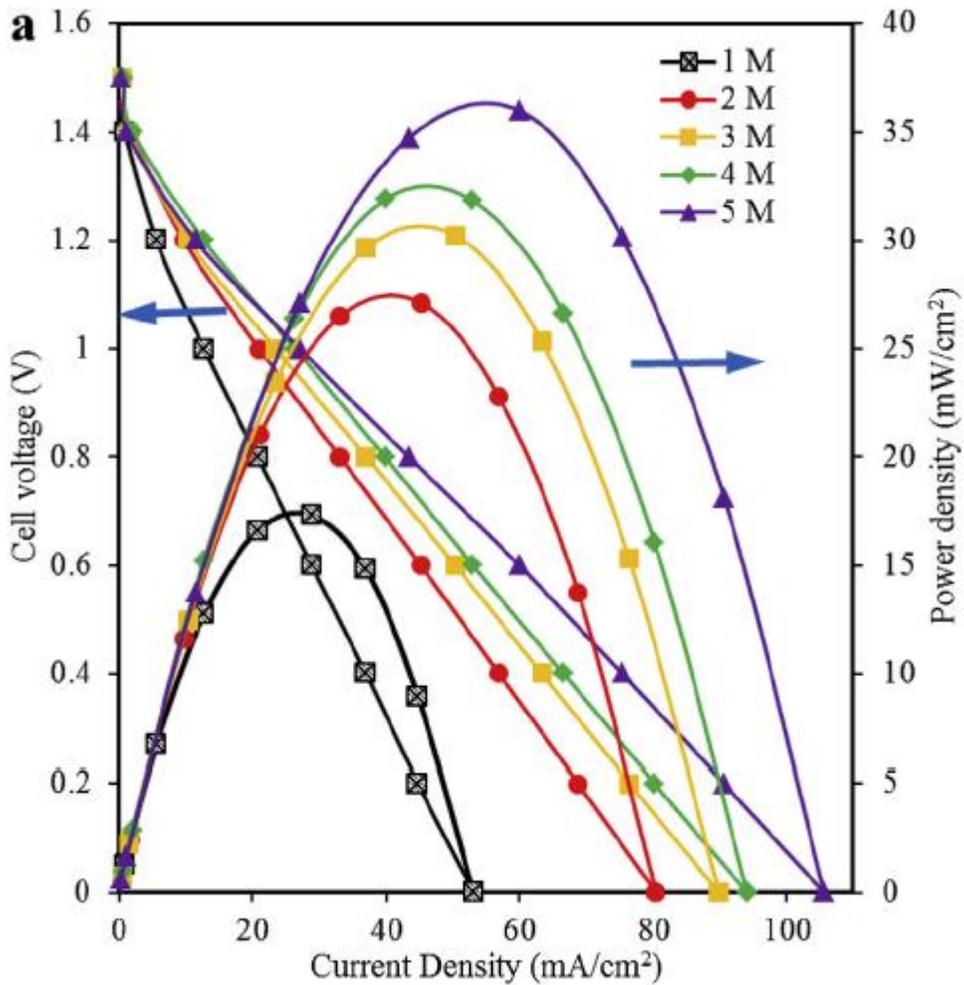


Figure 2-2 i-V polarization and power density curves (Lei Wang et al, 2013)

From figure 2-2, concentration of the electrolyte also have factor on the performance of the cell, as the molarity goes up the performance of the battery goes up.

Carbon dioxide in the air will cause carbonation of the alkaline electrolyte which can impede air access and cause harm to air cathode. When the saturation level for  $\text{Al(OH)}_4^-$  is reached,  $\text{Al(OH)}_3$  precipitate of the hydrargillite phase will be generated, The  $\text{Al(OH)}_3$  will binds to water, decreasing its accessibility of oxygen reduction at the cathode and it also block pores, this is worse especially for Al-air battery that use static electrolytes. The accumulation of the  $\text{Al(OH)}_4^-$  in static electrolyte will generate a viscous gel layer, the gel will slow down the diffusion of the  $\text{OH}^-$  ions towards the Al surface. This cause the rate of aluminium dissolution reduce. ( M.L. Doche et al. 1999)

To overcome the fluctuating electrolyte conductivity as well as the accumulation of  $\text{Al}(\text{OH})_3$  at the electrodes, the large scale aluminium- air system typically install a pump that connect to external reserve tank to circulate electrolyte. This prevent the building up of which lead to blocking of cathode pore. Heat exchanger and filter/ crystallizer unit can also be incorporated into the circulation of electrolyte cycle. Heat exchanger provide a thermal solution to control the temperature of electrolyte, stopping it from overheating and boiling. Where a crystallizer promotes the crystallization of  $\text{Al}(\text{OH})_4^-$  into the insoluble  $\text{Al}(\text{OH})_3$  which can be filter out, this will increase the  $\text{OH}^-$  ion concentration. (D.R. Egan et al. 2013). The rise in electrolytic conductivity of electrolyte enable a more stable steady cell potential difference, the consistence of conductivity of electrolyte through the whole duration of battery operation will improve the energy density of the battery cell.

The fluctuations in electrolyte conductivity in the beginning of battery operation can be avoid by adding  $\text{Al}(\text{OH})_3$  seeds crystals of typically  $1\mu\text{m}$  in size, into the electrolyte, The seed will cause saturation of aluminate. Aluminate ions formed will directly precipitate forming aluminium hydroxide which then can filtered out from the aqueous solution.(M. Nestoridi et al. 2009)

## 2.5 Summary

The aluminium air battery have few key component

- air cathode
- aluminium electrode
- electrolyte

The air cathode allow permeable for air and do not allow permeable of water, this allow the occurrence of ORR and is achieve by utilizing carbon based material with high surface area along with other component such as binder, charge collector and catalyst.

Aluminium is the main component used for aluminium cathode, aluminium alloy is preferred because parasitic corrosion can be reduced.

Alkaline electrolyte have higher conductivity, it also have hi  $\text{Al(OH)}_3$  solubility compared to saline electrolyte, this allow Al-air battery to achieve higher cell power and higher energy density, making alkaline solution the best candidate among the acidic, neutral and alkaline electrolyte.

Thus the aluminium air battery should consist of a carbon base air cathode, aluminium alloy as anode and alkaline solution as electrolyte.

## CHAPTER 3

### 3 Methodology and work plan

#### 3.1 Introduction

This section lay out the procedure and step required to be taken to achieve the objective of this project. The approached taken is justify in this section. Current design of aluminium air batteries is being review and intergrade into the existing car batteries design. A single cell and a multi cell battery is being design and the material, fabrication method and cost of the battery we be analyses. A structure work plan is being constructed to monitor the progress of the research. Information gathered from the literature review will be used to design and fabricate the aluminium air battery prototype. The flow chart (figure 3.1 shown below will be a reference guideline to carry out this project.

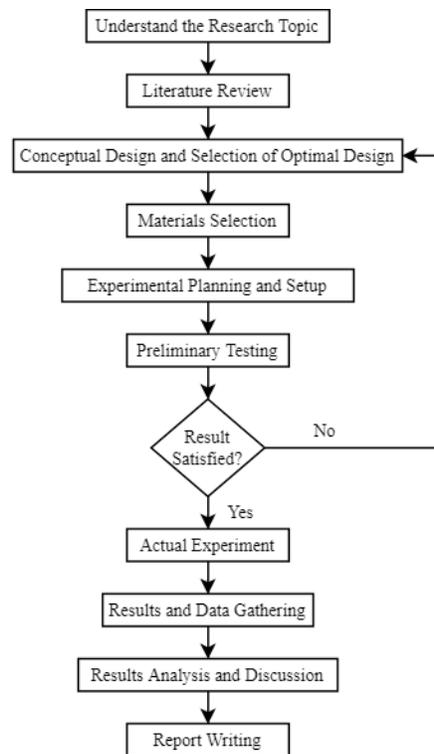


Figure 3-1 work flow of the project

### 3.2 Design the battery

The main component of the aluminium batteries design in this paper is mainly composed of four components,

1. Air cathode
2. Aluminium anode
3. Electrolyte
4. Container

Figure 3.2 and figure 3.3 show the structure of the batteries. The single cell is then connected in series to achieve higher voltage. The single cell is connected in series shown in figure 3-4.

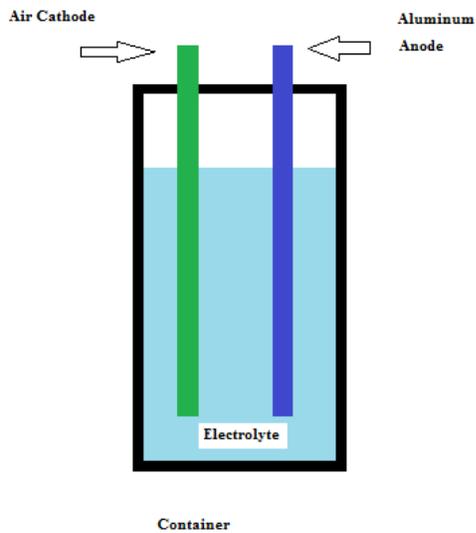


Figure 3-2 Simple design of the single cell aluminium air battery

The Al-Air cell should be able to function as long as the present of aluminium, Oxygen and water as shown in the primary overall reaction (1-3), therefore the minimum component required for this battery to function is to have an aluminium anode and air cathode submerge in electrolyte.



Figure 3-3 simple design of a single cell

A simple cell was built to show that the battery will function, consist of a aluminium anode made from aluminium foil, air cathode made from charcoal glue to stainless steel mesh and salt water as electrolyte (all these material can found in kitchen), this simple battery give off 1.1Voc and is show that aluminium air battery can be built with minimal component that is available.

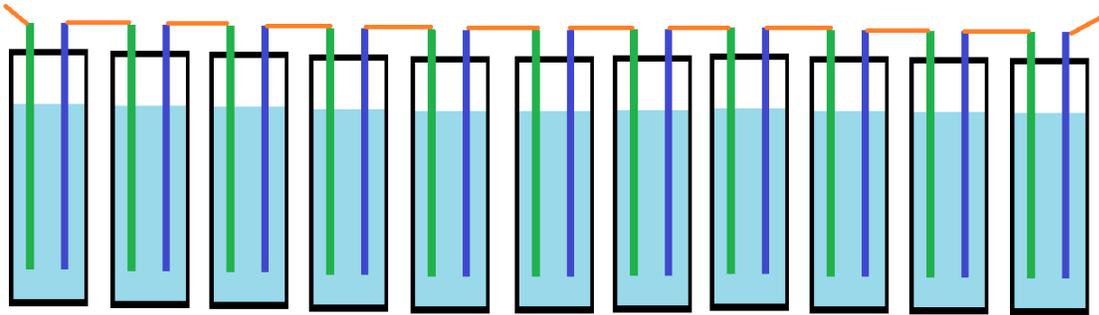


Figure 3-4 Multi cell aluminium air battery

To achieve higher voltage, single cell in connected in series mimicking the traditional acid battery used in car.

### 3.3 Initial design

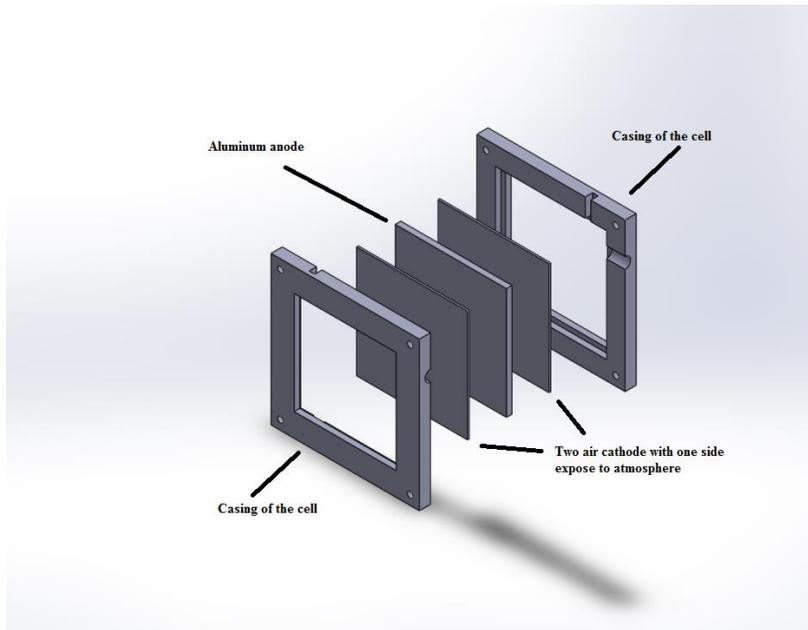


Figure 3-5 design of the proto type battery

The initial design is to build a single aluminium cell shown in figure 3-5 above consist of a casing that house two air cathode that house an aluminium anode in the middle. At the top of the cell there is two hole to allow wire to connect to anode and cathode. The housing consist of plate A and plate B.

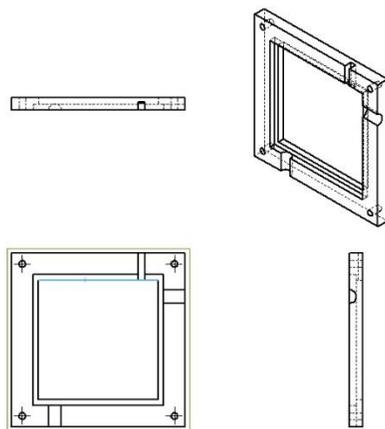


Figure 3-6 3D model of plate A design in solid work

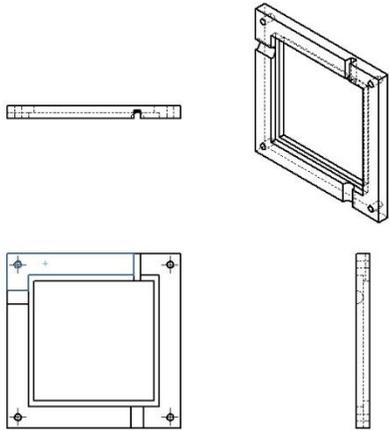


Figure 3-7 model of plate B design in solid work

To manufacture this two plate, few manufacturing method have been considered, traditionally it can be machine our from a block of the raw material such as acrylate block or metal block, the housing need to be nonconductive, so metal block is not suitable for the housing material.

Other than machining the structure out, the structure can be cast out from mold using different type of plastic as material, but to build a mold just for this project may not be financially viable as it is still a proto type and design changes will occur along the project.

3D printing come to the picture as it can print unique shape without required a mold and can print different material including many type of plastic and ceramic.

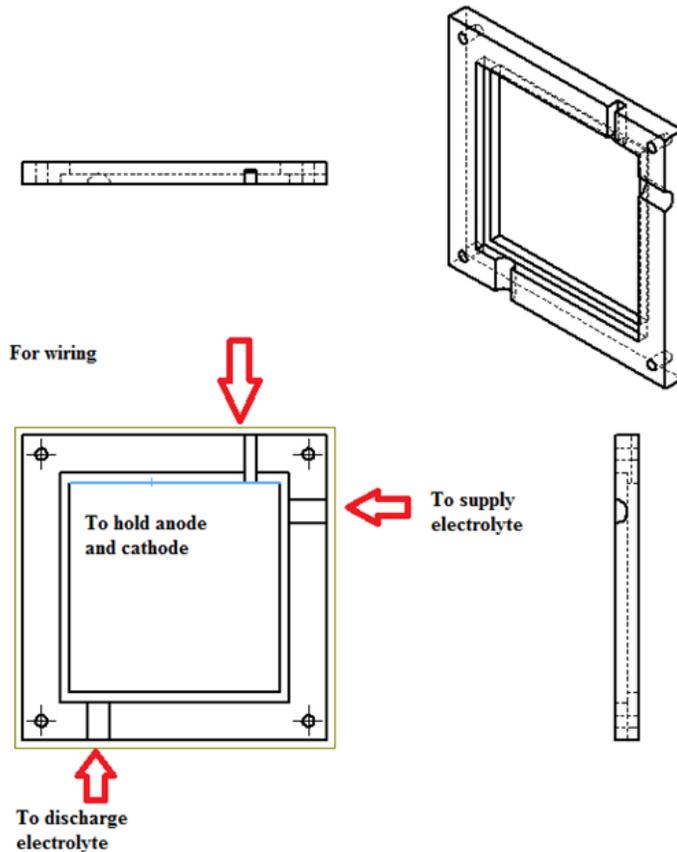


Figure 3-8 detail of the housing

The housing middle part of the casing which house the anode and cathode is design in a square/rectangle shape to have an easier manufacturing process for the cathode and anode material. This design also maximize the contact surface between the cathode and atmosphere.

The hole at located at four corner is for screw to screw all component together. To ensure it is water thigh, O-ring or gasket may be needed when screw in plate A and plate B together.

To achieve 12 V, is estimate that the system require 15 cell connected in series. Originally the design is to stack them next to each other, but one concern is that if the cell is stack too close, the battery cell may not have sufficient oxygen to react. So a 5 cm gap was introduce to each cell. But 15 cell stack together with 5cm gap is too long. So below arrangement was design.

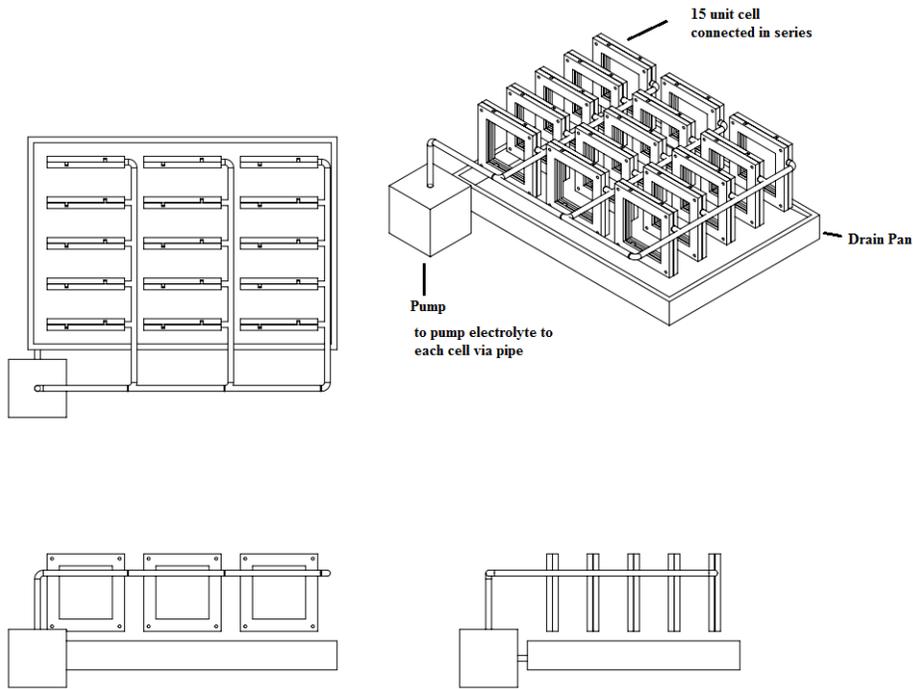


Figure 3-9 initial design of 15 cell stacking together

In this design a pump is introduced to deliver the electrolyte to each cell via a piping network. The discharge electrolyte is collected with a drain pan and connected back to the pump.

To further improve the dimension of the battery pack another configuration is designed.

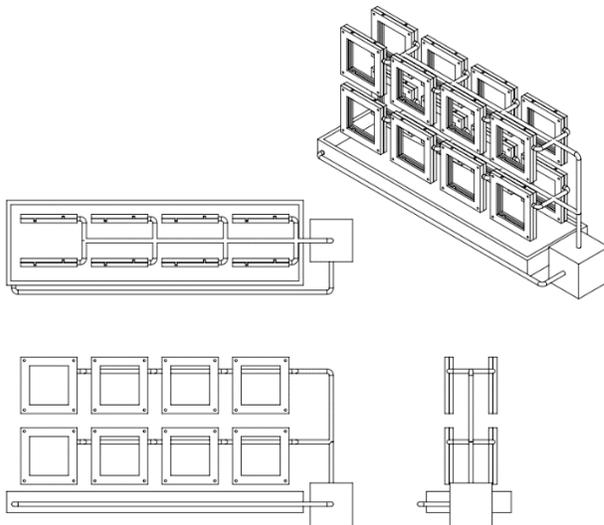


Figure 3-10 alternative arrangement of the cell

The design is send to a 3D printing supplier for quotation and the material selected for printing the structure is PETG and ABS, this two material was being selected because of their chemical resistivity, structure strength and electrical property (nonconductor). The price of PETG is more expensive than ABS but PETG only have a working temperature up to 70 degree Celsius so is not suitable for this application. To print the configuration in figure 3-9, the ABS will cost around RM700.00. And to stack the cell in such manner, an additional support structure need to be built to hold each cell and piping in place.

After reviewing both the configuration, the size of the structure is being reduce and simplify, instead of independent cell, multiple compartment is fused into a single housing structure, and the piping network seems over design and complicated thus piping system have been simplify.

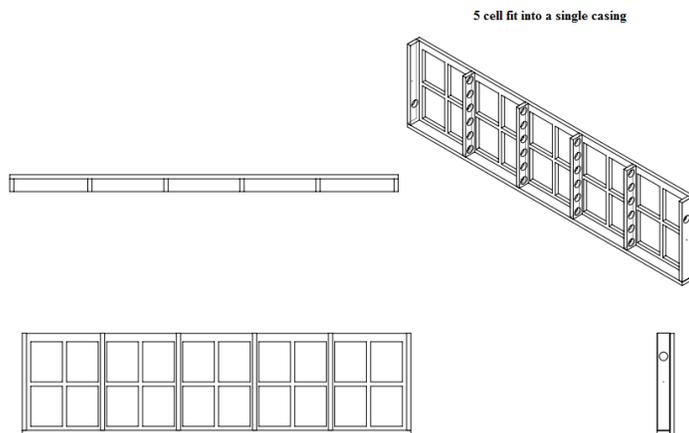


Figure 3-11 detail of multi cell housing

To overcome this, the cell is being redesign and the new design combine five cell into one casing. This will reduce the machining required for the casing material, and make the cell more compact. Each compartment will have hold a cathode and anode and will be connect in series to achieve higher voltage.

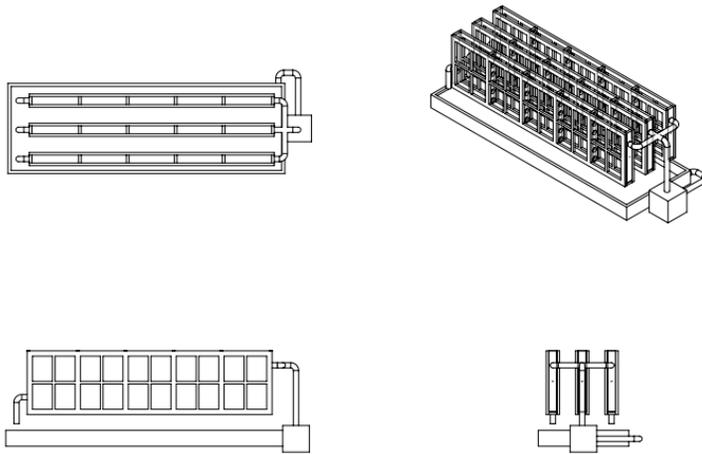


Figure 3-12 arrangement of multi cell housing

To further simplify the manufacturing process, 5 mm acrylate sheet is being used, and the structure of the housing from figure 3-11 is break into different plate. And the plate will glue together cold fusion via chloroform. The chloroform will dissolve acrylate into a liquid state and when the chloroform evaporate, the dissolved acrylate will turn into solid again, when chloroform is applied to two different surface of acrylate and pressing against each other, the diffused acrylate will make a new polymer bond and from a single structure, this required a smooth surface to maximize the contact area and this type of joining is consider a cold fusion.

A sheet of acrylate size 4ft x 8ft, 5mm thick cost RM 300, the required amount is 25% of that, which which cost RM 77, together with labor and machine required to process the acrylate, it will still cost mush less than RM700 (3D printing cost) thus the cost is also one of the driving factor for acrylate being used for this project.

### 3.4 Material Selection

Based on the literature reviews and the material availability the initial material for each component is selected as below

#### 3.4.1 Air cathode

The air cathode will be made by carbon base material as the large surface area of the carbon base material provide

Graphite block commonly used as electrode can be machine into different shape and is cheaper compare to carbon paper. It also have good electrical conductivity, pressure resistance, corrosion resistance, low expansion coefficient, good chemical resistance.

### **3.4.2 Aluminium anode**

99.5% purity aluminium (Young et-al, 2015), higher purity aluminium is quite expensive, they also corrode much faster compare to aluminium alloy because of this two factor a more common aluminium alloy is being used.

By alloying Mg, Zn, Pb, Sn, Ga, In, Mn, Hg and Tl, the parasitic corrosion can be suppress. (S. Z. El Abedin et al. 2004) (M. Nestoridi et al. 2009) (Smoljko et al. 2012) (D. R. Egan et al. 2013) The nominal composition of type 6061 aluminium is 97.9% Al, 0.6% Si, 1.0% Mg, 0.2%Cr, and 0.28% Cu.

6061 aluminium sheet is being used for this project as it's easily obtainable, relatively cheap and consist of Mg which help suppress the parasitic corrosion.

### **3.4.3 Electrolyte**

Alkaline electrolyte have higher conductivity, it also have hi  $\text{Al}(\text{OH})_3$  solubility compared to saline electrolyte, this allow Al-air battery to achieve higher cell power and higher energy density, making alkaline solution the best candidate among the acidic, neutral and alkaline electrolyte. The alkaline electrolyte used NaOH and KOH. Based on, NaOH have its peak electro conductivity at 4 mol dm<sup>-3</sup> in room temperature and KOH have its peak electro conductivity at 7 mol dm<sup>-3</sup> in room temperature.

### **3.4.4 Container**

The container needed to be non-electrical conductive so that the current generated will not leak, and can have chemical resistant as it needed to contain the NaOH or KOH used as electrolyte.

The manufacturing method needed to be consider, as the battery is a prototype, only few unit needed to be manufactured.

One method that comes to mind is 3D printing because it can produce special shape item in small batched, do not required a mold and it is one of the newer technology for manufacturing process. The material selected for 3D printing for this application is PETG and ABS. As they both have

good physical property and good chemical resistivity and both are not electrical conductor. But to rent a 3D printer is expensive, also the operating temperature of PETG is less than 70 degree Celsius thus is not suitable for this experiment as the reaction will generate a lot of heat.

The other method is to machine Polymethylmethacrylate- (acrylate). This material is available in block form and sheet form. Acrylate can be glue together using chloroform. The chloroform melt the acrylate and after the chloroform evaporate the soften acrylate fuse back together as one rigid body.

The edge of the acrylate need to be smooth when glue together because chloroform do not act as gap sealer. The possible gap was then seal with silicone sealant to further prevent leakage.

### **3.5 Summary of material selection**

Model in figure 3-12 is being used as my final design. Graphite block is used as air cathode, 6061 aluminium sheet is being use as anode, and acrylate sheet is being used as container. Alkaline electrode will be used, the two alkaline electrode selected is NaOH and KOH.

Cost to manufacture the battery is also put into consideration as the material selected is cheapest among the option considered, and also availability, as the COVID19 pandemic on going many raw material have problem in term of delivery lead time. The material need to be send in in time for the project period for the system to be fabricate.

In the final design, two air cathode is surrounding the aluminium anode to maximize the occurrence of OOR by maximizing the surface area of the air cathode that can be expose to atmosphere.

## CHAPTER 4

### 4 Result and Discussion

#### 4.1 Fabrication of battery body

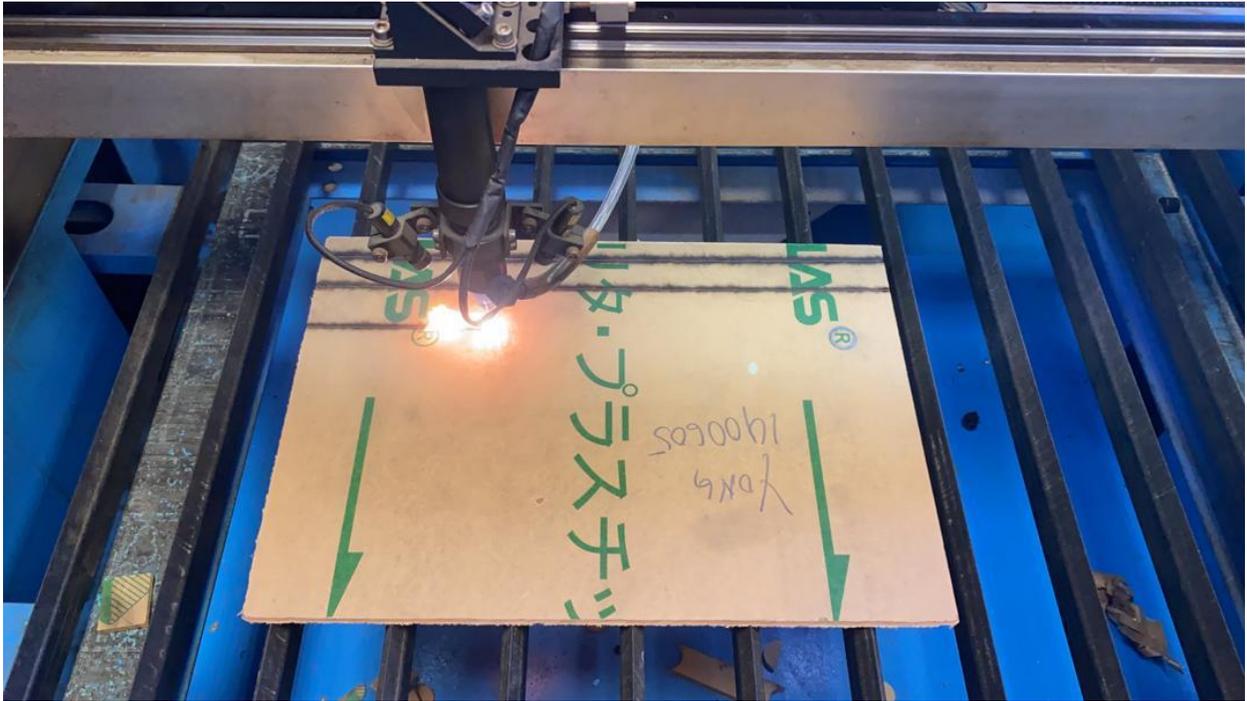


Figure 4-1 laser cutting acrylate sheet

The acrylate sheet is cut by laser cutter. Laser cutter is used because of the higher precision and better surface finish, smoother edge compare to machining.



Figure 4-2 acrylic sheet that finish cut by laser cutter

After the acrylic is cut, the quantity of each piece is checked. 2 additional spare piece was cut. Because of heat generated by the laser cutting smaller piece is curve after cut. The dimension of the piece is readjusted so that the piece is thick enough so that the cutting process will not curve the piece.



Figure 4-3 try fitting and dimension checking by ruler

The dimension of the cut piece is checked by ruler. The structure of the housing is design to be exactly 100mm x 50mm, and the graphite is also cut to size of 100mm x 50mm x 2mm, But the

laser cutting introduce heat to the 5mm thick acrylate sheet, for some of the smaller pieces, because of the heat introduces the sheet have deform into a slight curve sheet instead of a flat sheet. Additional stiffener made from acrylate was added into the structure.



Figure 4-4 cleaning and surface preparation before joining of the acrylate sheet

The protection film is remove and each piece is clean by thinner to ensure the surface is free of foreign object and ready for joining.



Figure 4-5 join fusion of acrylate sheet by using chloroform

The piece is try fitted and glue together with chloroform. During the gluing process, the curve piece is hold together by hand with sufficient pressure to ensure the cold fusion is complete before letting go to ensure the structure is cured according to desired shape. If multiple cell needed to be build this way, I will build a jig to fit in the piece before gluing them together.



Figure 4-6 try fit of machined graphite sheet into fused acrylic housing

Although the cutting of the graphite sheet was relatively uniform and accurate, when measured by metal ruler the size have a  $\pm 1$ mm in size tolerance. But because of the slight bend acrylic and the gluing process, the compartment design to hold the graphite is not a perfect fit. The graphite sheet in cut and sand down according to the case and try fit into the pocket.



Figure 4-7 application of silicone sealant to hold graphite sheet in place

Silicone sealant is used to hold the graphite sheet in place and prevent leakage, it is also use to fill the corner of the container to fill in the gap that may have between the acrylate sheets.



Figure 4-8 curing of the housing

After silicone is being apply the set is allow for curing for 24 hours.

## 4.2 Preparation of electrolyte

The liquid electrolyte is prepared by mixing NaOH salt with distilled water and KOH salt with distilled water according to the molarity required by mass to water ratio.

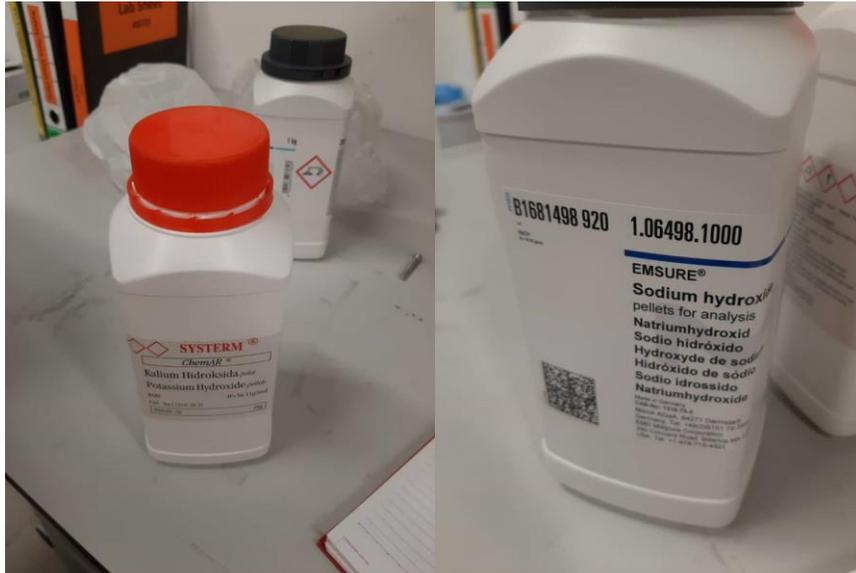


Figure 4-9 NaOH and KOH salt pallet

The molar mass of the NaOH and KOH is labeled on the salt pallet container. The molar mass of NaOH is 39.997g/mol and molar mass for KOH is 56.1056g/mol.



Figure 4-10 measuring NaOH in electrical scale with chamber to prevent air flow.

The pallet is measured by an electrical scale with enclosure. 79.994g of NaOH salt pallet was mix with 500ml of distilled water in a biker with a glass rod to prepare the  $4\text{mol/dm}^3$  NaOH electrolyte.

Although  $7\text{mol/dm}^3$  of KOH should have the peak electrolytic conductivity for KOH in room temperature, but  $7\text{mol/dm}^3$  is too corrosive and can be very hazardous to handle, a  $4\text{mol/dm}^3$  was prepared instead.

The  $4\text{mol/dm}^3$  KOH is prepared by measuring 122.211g of KOH salt pallet and mix with 500ml distilled water in a biker with glass rod.



Figure 4-11 Mixing of NaOH in biker with glass rod

### 4.3 Testing

1. Leak test
2. Polarization test
3. Constant current discharge test



Figure 4-11 leak test

Leak test was performed for 1 hour, to check if the cell is able to hold liquid water.

The cell is connected in series shown in figure below, and connected to VersaSTAT 4 Potentiostat Galvanostat to perform the polarization test and discharge test.

For polarization test, when the battery is on a steady state, the meter will draw the current starting from 0A with the increment of 0.00005A/s and the corresponding voltage is being recorded. The reading will stop once the Voltage drop to 0V.

For the discharge test is set to operate at a constant current of 0.05A then for every 1 second a measurement of battery Voltage is recorded until the battery Voltage drop to 0V.

The highest voltage is being recorded, the initial and final mass of the aluminium is being recorded. The data collected by the potentiostat is being plot into polarization curve and discharge curve.

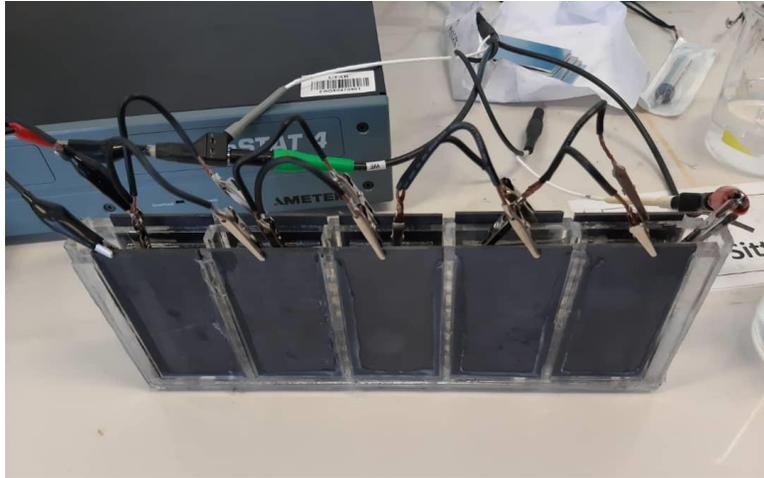


Figure 4-12 connecting aluminium anode to the graphite air cathode with alligator clip and 1.5mm<sup>2</sup> copper cable



Figure 4-13 after load the cell with electrolyte connected the cell with VersaSTAT 4 Potentiostat Galvanostat, bubble form during the test, may cause by formation of hydrogen gas or boiling of electrolyte or both.

During the test, few thing happened, after about 1min into the experiment, the electrolyte started to form a lot of bubble and vapor, the leakage of the battery is very bad and the battery got very hot.



Figure 4-14 sealant fail during test

The battery is then operate using  $4 \text{ mol dm}^{-3}$  KOH and  $4 \text{ mol dm}^{-3}$  NaOH

After 5 min the battery fail due to mechanically failure.

All 3 set of the battery fail within the 15 min. We reduce the molarity of the electrolyte into 1mol although the battery last longer but the battery eventually fail at 15 min into the experiment.

Constant current discharge test was unable to be preform due to structure failure of the battery.

The failure of the sealant may due to

- 1) Heat
- 2) Corrosion form electrolyte
- 3) Pressure generate by hydrogen

To improve the resistance against this three parameter, different type of sealant is being use and additional heat resistance gasket is being used.



Figure 4-15 type of high spec sealant and gasket used

A high performance epoxy sealant combine with a heat resistance gasket is being apply to secure the graphite sheet.



Figure 4-16 application of gasket

Before the application of the new sealant, the old sealant is being scrape off and the acrylate structure is being clean with thinner. The cleaning is important as it made a clean surface for the new sealant and gasket to be applied on.

The gasket is applied in the inner side of the graphite. The gasket providing an inner seal that can endure heat. The graphite sheet is then press into the liquid form gasket and allow 2 hours for the gasket to harden into solid form.

After additional sealant is being applied on the external of the graphite, the gasket should be able to provide additional sealing capability for the structure.



Figure 4-17 application of epoxy sealant on the graphite with gasket beneath.

Then the epoxy sealant is apply outside of the graphite to secure the graphite sheet and gasket into the casing. The epoxy sealant need one part epoxy and one part hardener, after the two component is mix well, the allowable working time is 20 min and the curing time is 24 hours.

After the epoxy sealant is harden. The battery is again undergo leak test for 1 hours with water to ensure there is no leakage under normal circumstance.

The battery is then connect to the VersaSTAT 4 Potentiostat Galvanostat and run the polarization test and constant current discharge test again.

The electrolyte used for the improve version of battery is NaOH 1mol and KOH 1mol. Although 4mol NaOH and 7mol KOH may have optimal conductivity but from previous experiment, the corrosively of the high molarity electrolyte may damage the sealant. The higher reaction speed also cause heat generation and high volume of H<sub>2</sub> gas formation.

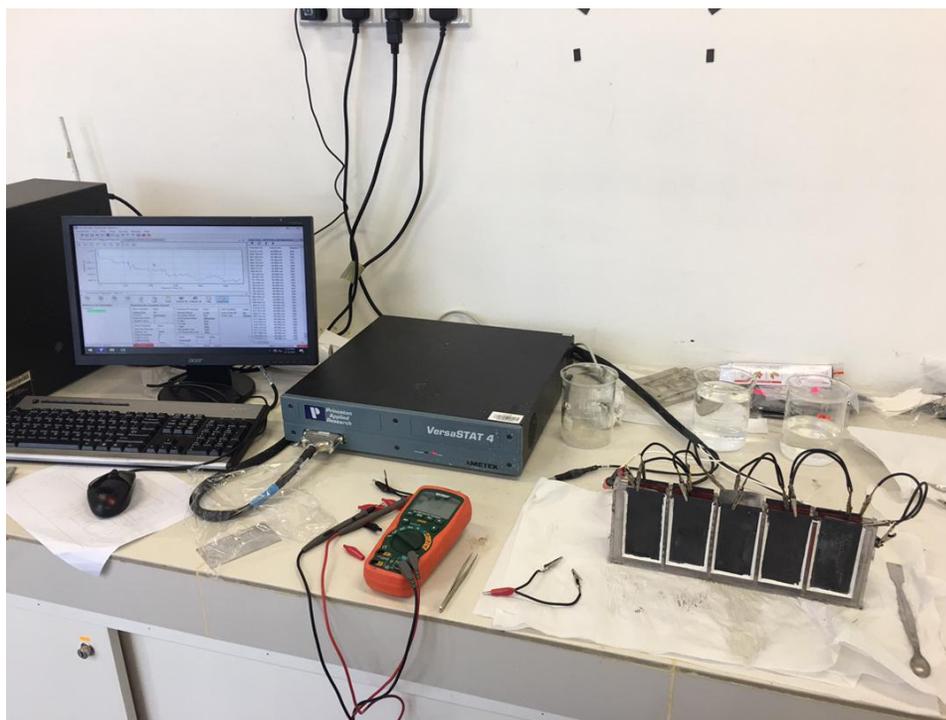


Figure 4-18 battery is set up and connect to VersaSTAT 4 Potentiostat Galvanostat to preform test. The voltage get from the single “5” cell battery is 1.2V, which is much lower than what I expected, then after some reach. I realize that the configuration what I am doing is a battery with short circuit, as they share the electrolyte, the battery act as 1 battery cell.

After the 2 set of test result of this set up with 2 different electrolyte, I re-configure the cell into a 3 separate cell and connect them in series and run a polarization test. The new configuration shown in figure 5-9 below are able to obtain a Voc of 3.3V, which confirm that each of my battery cell have a voltage around 1.1V to 1.2V and if connect in series, higher voltage is able to achieve.

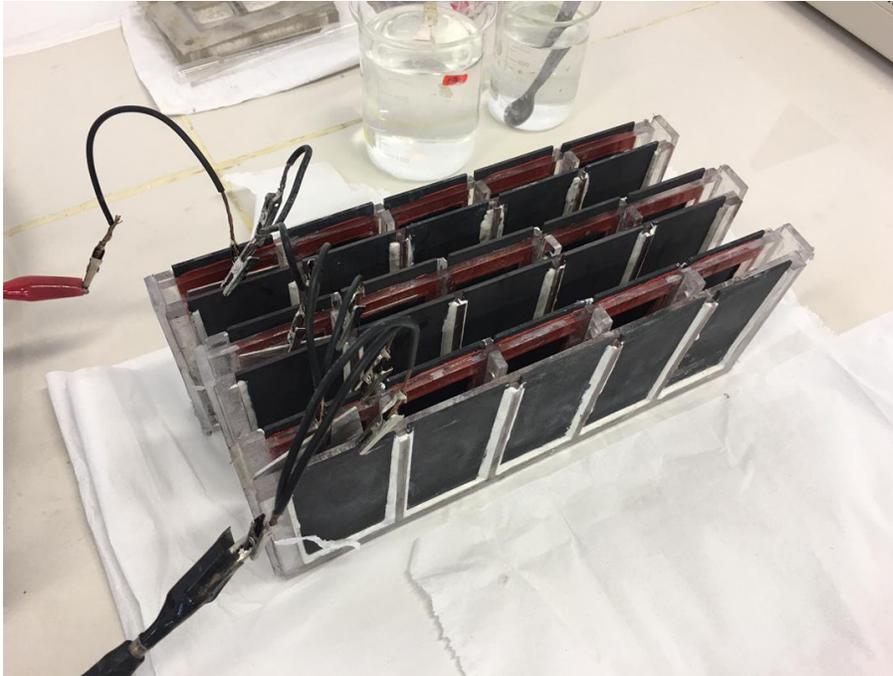


Figure 4-19 Battery is set up in a 3 cell in series.

After the test, the electrolyte is again collected into a beaker and the electrolytic conductivity of the electrolyte is being measured using electrical conductivity meter.

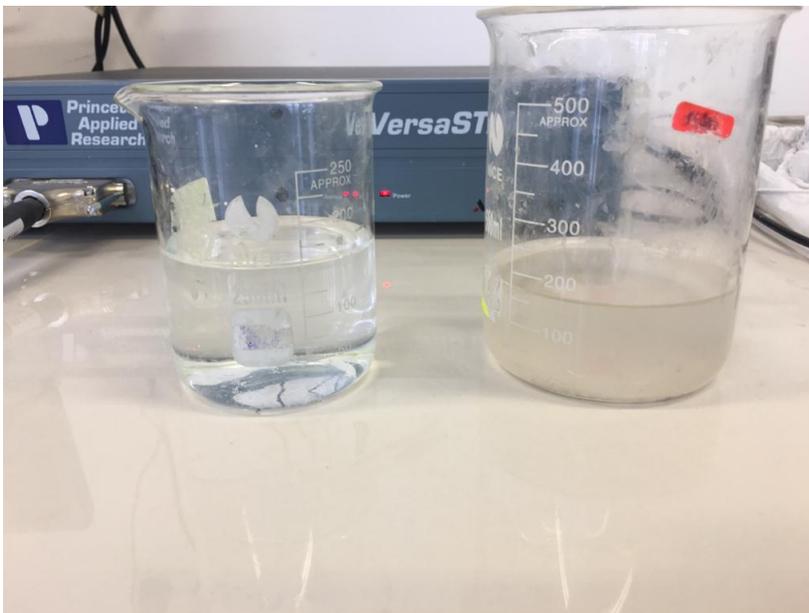


Figure 4-20 electrolyte before (left) and after (right) the discharge test

From the picture, you can see that the electrolyte turn cloudy in color and the viscosity of the electrolyte greatly increase into a gel like texture. The conductivity of the electrolyte also drop after the discharge test as shown in figure 5-11 and 5-12.

This is cause by the formation of  $\text{Al(OH)}_3$  that dissolve in water,



Figure 4-21 Electrical conductivity of  $\text{KOH } 1\text{mol/dm}^3$  before the discharge test, measured using electrical conductivity meter

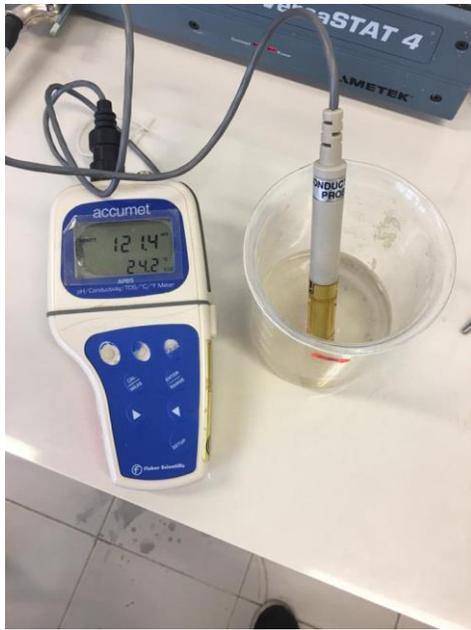


Figure 4-22 Electrical conductivity of KOH 1mol/dm<sup>3</sup> after the discharge test, measured using electrical conductivity meter

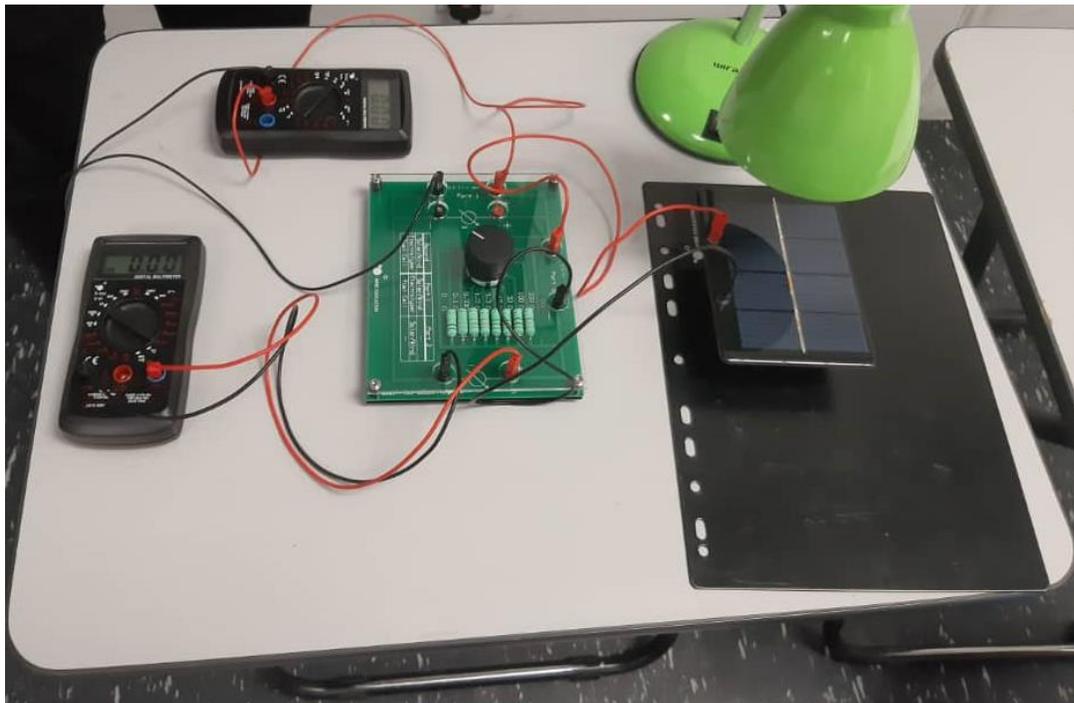
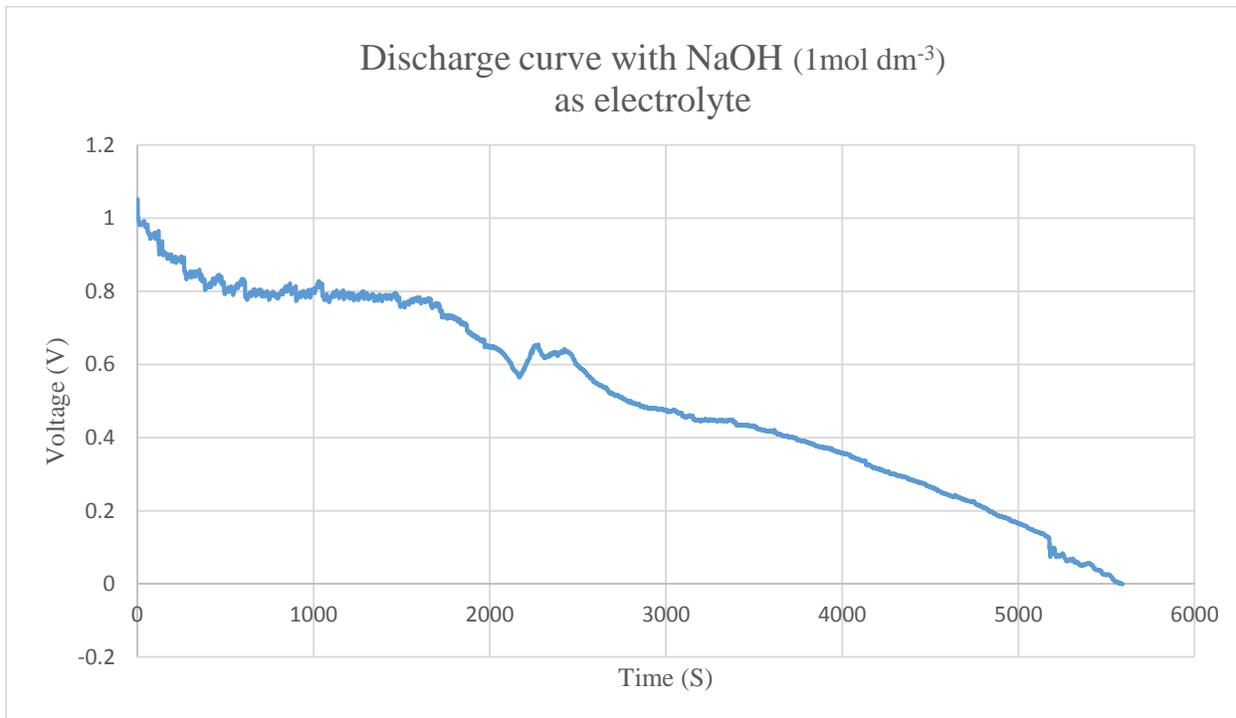
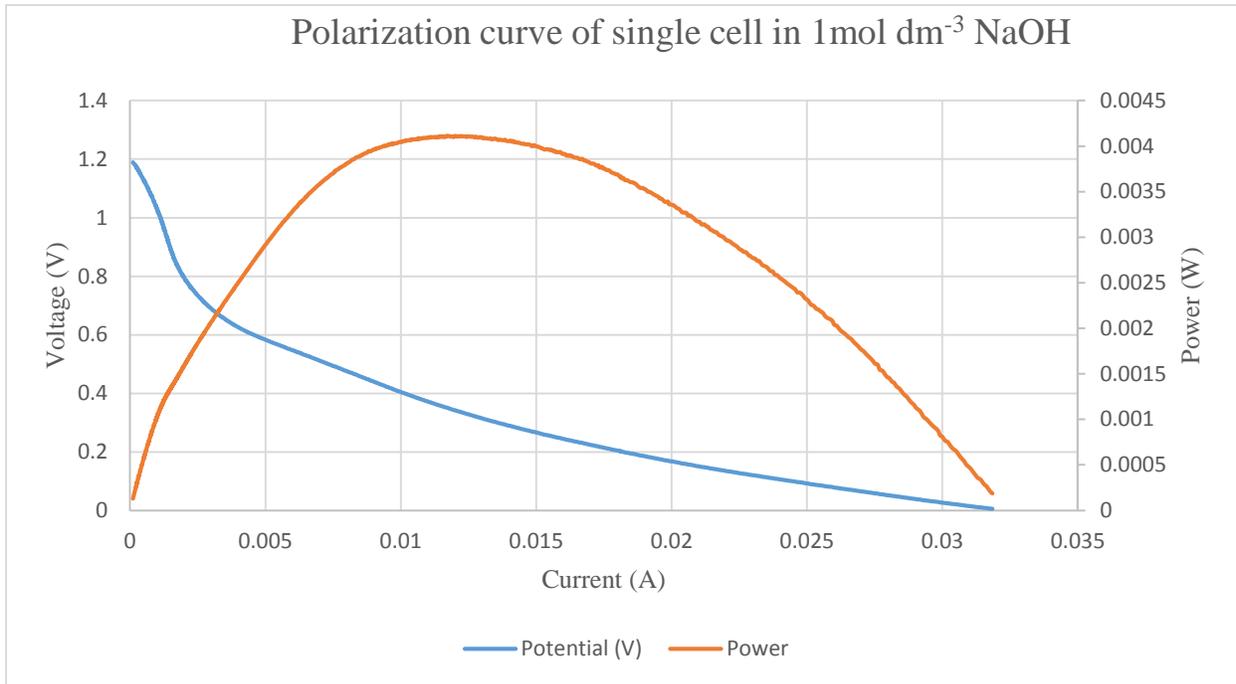


Figure 4-23 alternative method of measuring the performance of the battery.

A more simple approach to measure the performance of the cell can be done by connecting the battery to a series of resistor and measure the voltage and current across the resistor with voltmeter and ammeter.

#### 4.4 Result

The first set of data is gathered from single cell aluminium air battery with  $1\text{ mol dm}^{-3}$  as electrolyte



The discharge current is set to be 0.05A and the voltage and the corresponding Voltage is being recorded, as shown in the graph above, the battery run for 1.55hr (5592 sec) until the voltage drop to 0V.

The starting mass of aluminium sheet is 16.647g and after the discharge test the mass of aluminium is 14.338g, the lost in mass of aluminium is 2.3g. from figure 6-1 we can see that most the aluminium still intact.

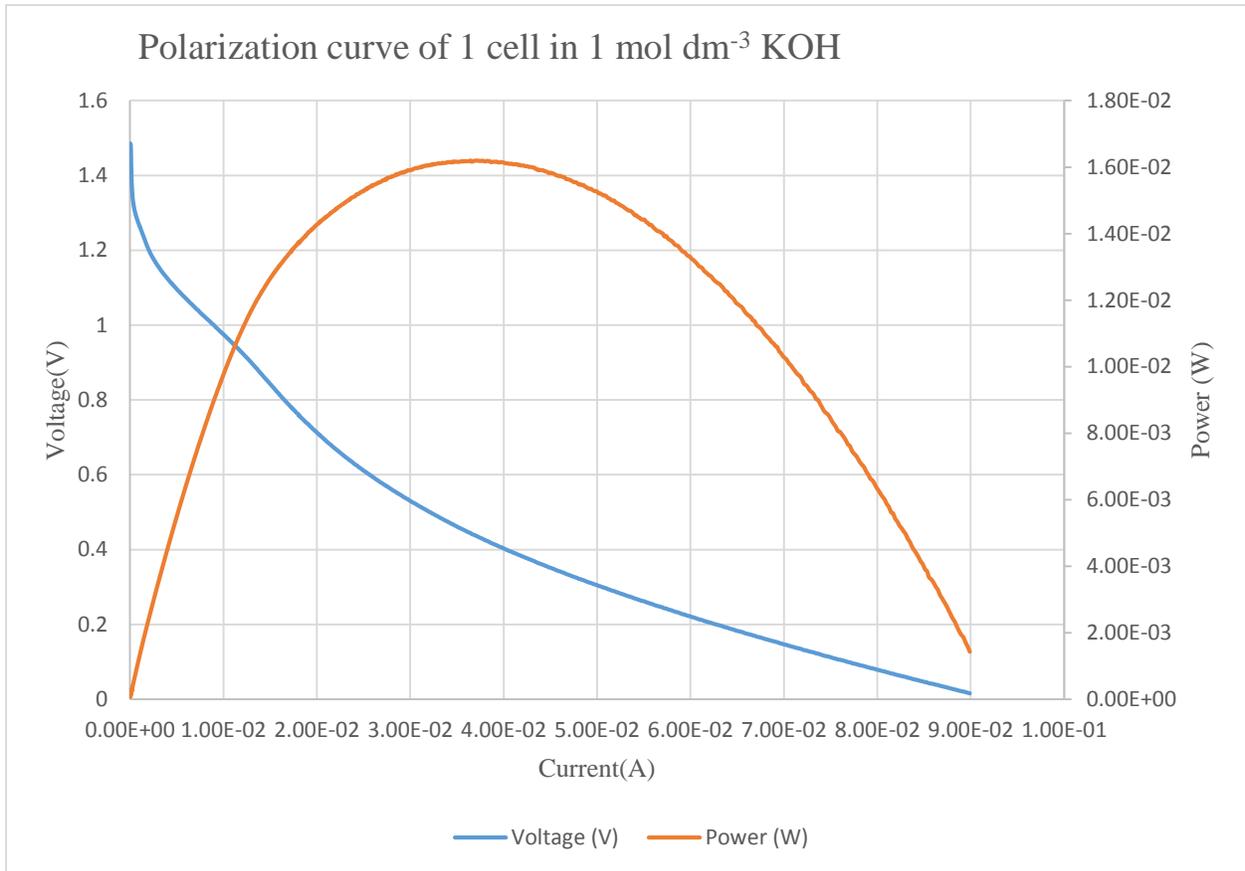


Figure 4-20 aluminium sheet after discharge test

Charge release =77.652mAh

Max power =0.004116W

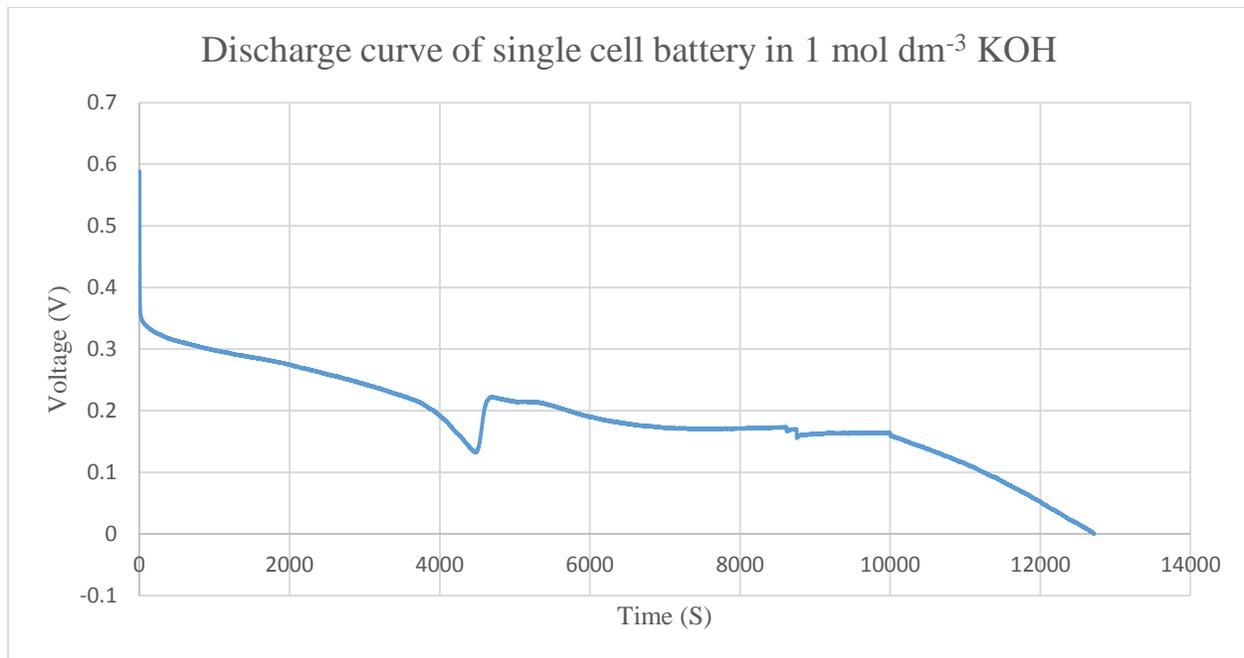
Then the electrolyte of the battery is change to 1 mol dm<sup>-3</sup> KOH to compare the performance of the battery in different electrolyte.



A higher Voc, more current and more power is able to draw from the batter in KOH electrolyte.

$$\text{Max Power} = 0.0162\text{W}$$

KOH have higher solubility for oxygen, higher oxygen diffusion coefficient and lower viscosity. The solubility limit of the reaction product aluminate is higher for KOH than NaOH. (D.R. Egan et al. 2013) Thus the KOH electrolyte battery perform better than the NaOH electrolyte battery.

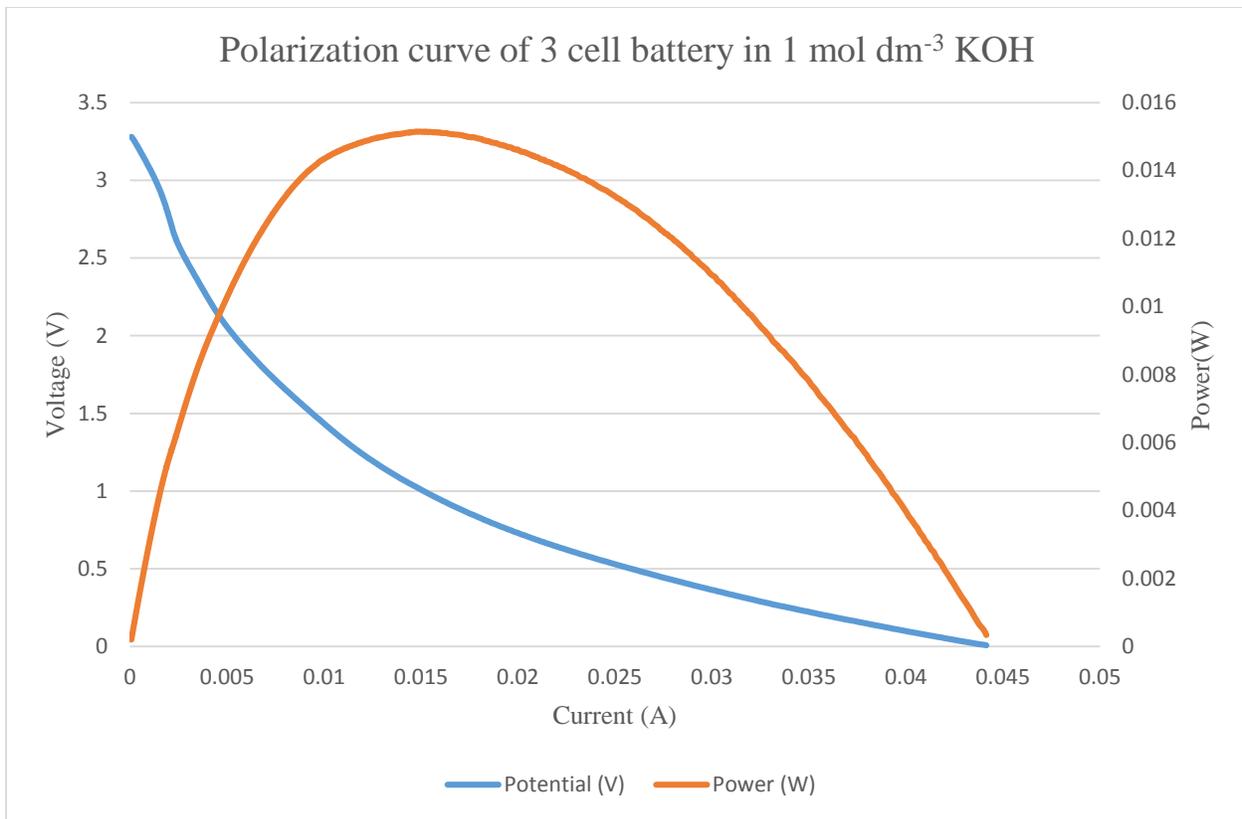


The discharge current is set to be 0.05A and the voltage and the corresponding Voltage is being recorded, as shown in the graph above, the battery run for 3.53hr (12717 sec) until the voltage drop to 0V.

The starting mass of aluminium sheet is 18.603g and after the discharge test the mass of aluminium is 15.483g, the lost in mass of aluminium is 3.12g. Voc recorded is 1.231V.

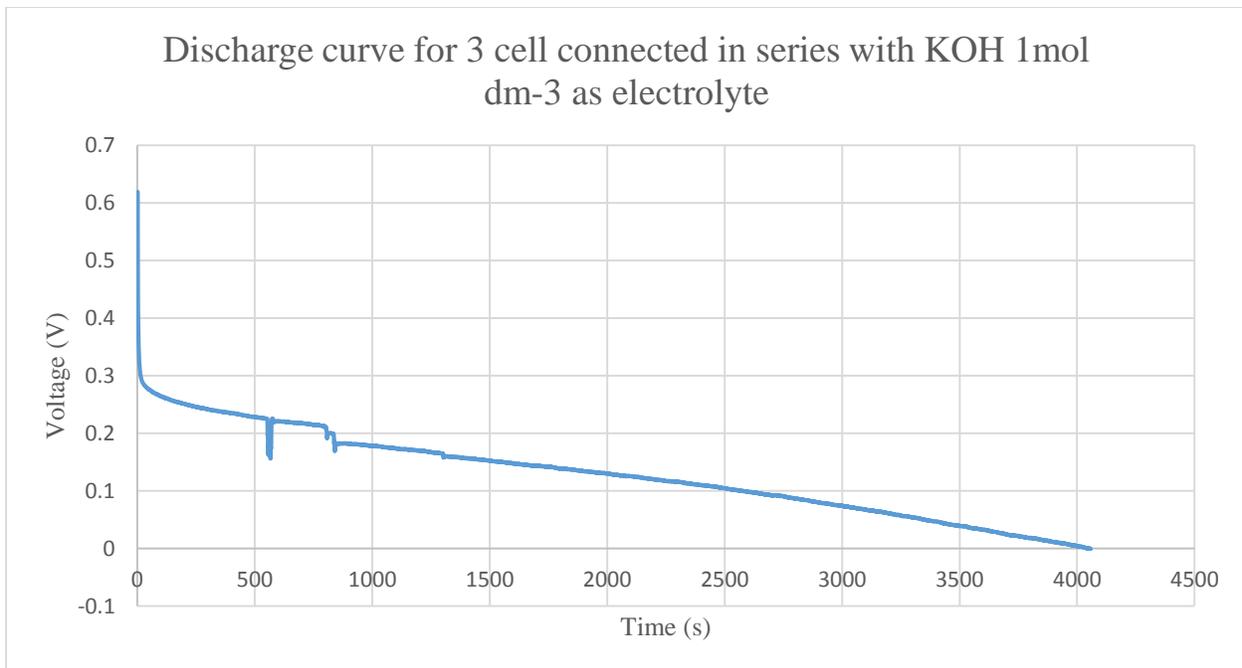
Charge release = 176.6111 mAh

In this discharge curve test, during the 4000s to 4300s, there is a voltage drop, this maybe cause by the inconsistency of the electrolyte as the property of electrolyte will change trough the span of the experiment due to the formation of Aluminium hydroxide that mix with water.



As shown in figure 5-9 the 3 cell set up is connected and polarization of the 3 cell is conducted. The Voc is 3.28V and the maximum power out is 0.01515W. The power output is similar to the single cell set up May due to the reduce amount of the aluminium used as I only put in 3 aluminium plate to conduct the 3 cell in series test.

There is a significant raise in open circuit Voltage, there the power generated is not much higher than a single cell battery that operate in KOH 1mol dm<sup>-3</sup>.



The discharge current is set to be 0.05A and the voltage and the corresponding Voltage is being recorded, as shown in the graph above, the battery run for 1.27hr (4060 sec) until the voltage drop to 0V.

The starting mass of aluminium sheet is 45.325g and after the discharge test the mass of aluminium is 43.079g, the lost in mass of aluminium is 2.246g. Voc recorded is 3.177V.

Charge release = 56.3611 mAh

It is found that the loss of weight in aluminium is around 5g on average per cell, for the same amount of electrolyte, the battery should use a 10g aluminium sheet to reduce the weight of the battery as anything more than 10g will not be react.

## CHAPTER 5

### 5 Conclusion and future plan

A multi cell battery is been build and being analyze, feature of the battery is being explore and through the process, some characteristic of the battery is being confirm and plan to be improve. Which although the performance of the battery not as good as battery that have been built by (Lei Wang et al, 2013) Wang which can achieve  $45\text{mWcm}^{-2}$  or (Zu Liu et al, 2016) Zu Lius battery which the battery achieve 80A with 120W.

Some aspect of the battery that was not covered in the literature review, which have bought to attention during the construction and testing of the battery. Two of the factor is heat management and chemical resistivity of the sealant. Form the first experiment, the case using silicon base sealant have no problem holding tap water for one hour but during the leak test, all the sealant fail to contain the electrolyte and started to peel off after the electrolyte been fill in and the electrochemical process start, and once heat buildup in the. After change to epoxy sealant, the leakage problem have been solved.

There is many research on each component of the battery, the material selected for each component is based on the previous research done, this project is to optimize the battery design and fabricate the battery base on the design. Other than the structure of the battery, the open circuit voltage and the current generated need to be improve because compare to previous study, this battery have low voltage and current.

The share electrolyte for the five compartment with the original wiring shown in figure 4-12 causes short circuit and give off unstable reading during the first test, the configuration of wiring which was then changed to figure 4-19, but in this configuration, eight of the air cathode in each cell was not connected to any wire which may be the reason of the low current that the battery give.

Although the open circuit voltage can achieve 1.2V, but my battery can only produce 0.03 to 0.05A with maximum power output of 0.015W, which is really low. The other problem arise after is the release of hydrogen gas during the experiment. A hydrogen gas collection system will be a good function to add into the battery cell, as hydrogen gas can be used to power fuel cell.

This project set a base on how to build a simple aluminium air battery, and from this base the battery can be further improve the design by improving the structure of the battery, material used for each component, wire connection of the battery. Additional component such as charge collector, circulating electrolyte, H<sub>2</sub> gas collector, heat management system and catalyst can be added into the system to further improve the power and energy generated by the battery.

The performance of the battery can be improve by adding charge collector to better collect the charge and improve the current generated, and also include a circulation of electrolyte to give a better electrical and chemical property of the electrolyte. The circulation of electrolyte can also provide strong alkaline that can dissolve the aluminium protective layer from and will have less saturation of Al(OH)<sub>3</sub> in the fresh electrolyte that been pump into the system.

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