Development of a maintenance free tropicalized lead acid battery enhanced by membrane technology

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Abstract

The objective of this study is carried out to develop a Maintenance Free Tropicalized Lead Acid Battery enhanced by membrane technology, to overcome problems such as lost of electrolyte, spilt of battery acid onto the battery compartment, and other associated enviamential problems. Asymmetric gas separation membranes were fabricated using polysulfone polymer and solvent. Polymer and solvent concentration were varied with the range of 9.5% to 30% and 90.5% to 70% respectively. The produced membranes were then incorporated into the Lead Acid Battery vent holes that have been designed in this study. Results showed that when the battery was charged at 13.2V (2.2V per cell) over period of 168 hours at 26°C ± 2°C the electrolyte losses is indistinctive, but with charging rate at 14.4V (used by Proton Car) the loss of electrolyte is fall below permitable level with the same parameter. Heat simulation test was concurrently carried out at temperature of 26°C ± 2°C over period of 168 hours without charging, the electrolyte loss was undistinguished but when the battery was simulated under hot bath condition at 82°C ± 2°C the loss is 1.6kg over the initial weight of 12.8kg.

Keywords: Lead acid battery, electrolyte, charging/discharging reaction, polysulfone gas separation.

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1. Introduction

Lead-acid, the most common storage batteries in the world are the lead-acid batteries used to start automobiles, UPS and others which are not only the heirs to Plante's first inventions but almost identical[1]. They have anodes made from porous lead and cathodes made from lead oxide, both soaked in a sulphuric acid electrolyte.

On the other hand, besides being cheap, lead-acid batteries have over 150 years of technical development behind them. They can be custom-tailored to specific applications, such those requiring deep discharge cycles. For example, where the batteries are used as the sole power source for electrical equipment, or for battery backup uses such as in large uninterruptible power supply systems(UPS) in data centres. Lead-acid cells also have a low internal resistance and thus can produce enormous currents and also good efficiencies of power to weight ratio (Wh/Kg) yet they suffer no memory effect. The cells also have a moderately long, predictable life. And of course, they are cheap[8].

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The Importance of the Lead-Acid storages Battery. An intelligent and the lasting battery is the ultimate desire of most drivers. Almost every function in today's

automobile depends on electricity. These include the electric windows, mirrors, seat adjustment, adaptive brake, central lock, security system, adaptive steering and most important; the Engine Management System (ECU). Of course, each and every feature plays an important role of the controls, while driver orientated are emphasis on the enhancement of safety, in order for the driver to focus his full attention on the road.
However, this increasing demand of the modern technology required more source of adequate power to support this newly enhanced and improved standard of comfort, safety and conveniences. Thus, the importance of the lead acid battery is specially designed to get through this severe demand in this competitive edge market. The lead acid battery has a proof -of-competency and reliability records during the testing and is guaranteed a long life whether in charging or discharging.

The lead acid battery especially in tropical countries and countries which has hot summer has critical Electrolyte decomposition problem that has deteriorating its life cycle and performance. The combination factors of high rate equalising charging at 14.4V to 15.8V, and hot weather contribute to the electrolyte losses in the lead acid battery.
This wasn't an arduous task, it only entailed removing the caps of each cell and distilled was added as needs. But it messy and was often bypassed or overlooked by drivers Whenever, the electrolyte level goes down below the minimum level, both positive and negative electrodes plates are exposed to air, thus sulphurtion takes place and the battery will be permanent damaged. As for tropical climates and countries with high ambient temperatures the predicament of exploding batteries is particularly acute.

Some manufactures introduce the so-called sealed maintenance free battery (SMF) that uses Gel Electrolyte in theirs products. However, it is a disappointment that this type of battery is good to operate at the ambient temperature not excess of 48 ºC These types of batteries were imported and have been sold in our country, but unfortunate, the batteries perform very short life, due to in-mobilised gelled electrolyte and plates, when temperature is above 48 ºC.

The second type of the maintenance free batteries has a valve inbuilt to regulate pressure generated from the cells. This type of battery so-called valve regulated lead acid batteries or (VRLA). With the valve inbuilt, the common ventilation hole is closed at all time until the internal pressure built-up to 0.3 bars, than opened released for a short time interval of about 1 to 3 second, at the temperature of about 35ºC. With this type of design, it works fine if the operation temperature is not exceeding 48 ºC. Typical problem of these batteries is the malfunction of the regulating valves that cause the batteries to swell or the top cover to rip-off, and electrolyte spilled out from the battery cells as shown in Figure 1.

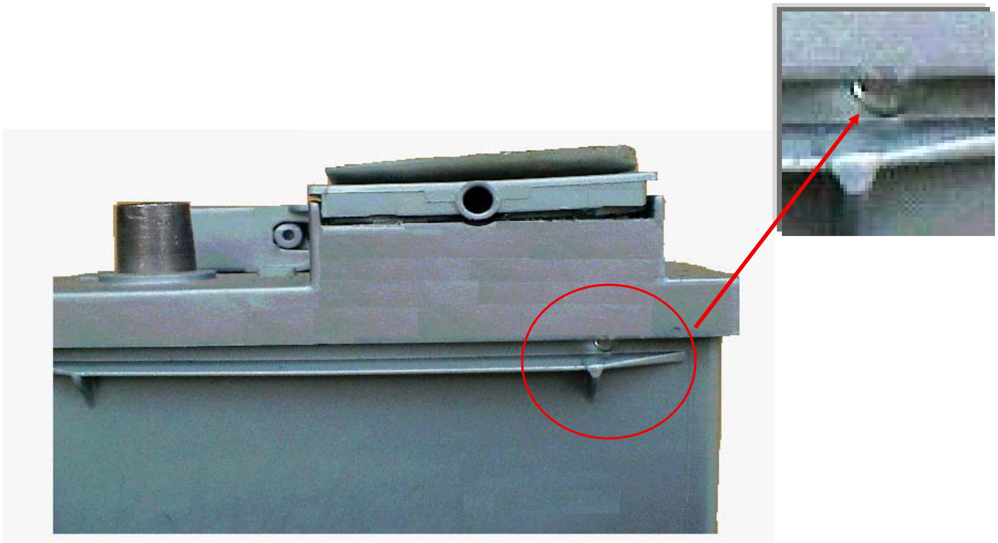

Figure 1: Electrolyte escape from the cells of a valve regulated type battery

It is essentially that the battery must design to suits the tropicalized climate. The reasons are to prevent the followings, which happened very frequent particularly in the tropical countries such as Malaysia, Thailand, Indonesia, Philippine and or countries which have long and hot summers.

a) Gaseous escape
b) Lost of electrolyte
c) Acid spilt
d) Corrosive to battery compartment and vehicle body

Actually, acid with lead from the cells has been discharged through the ventilation hole, and indirectly it will be discharged to the environment, thus becomes environmental unfriendly. Usually, the discharge contains lead acid that will cause permanent damage to metal parts and toxic to leaving creatures as well. Example shown in Figure 2.

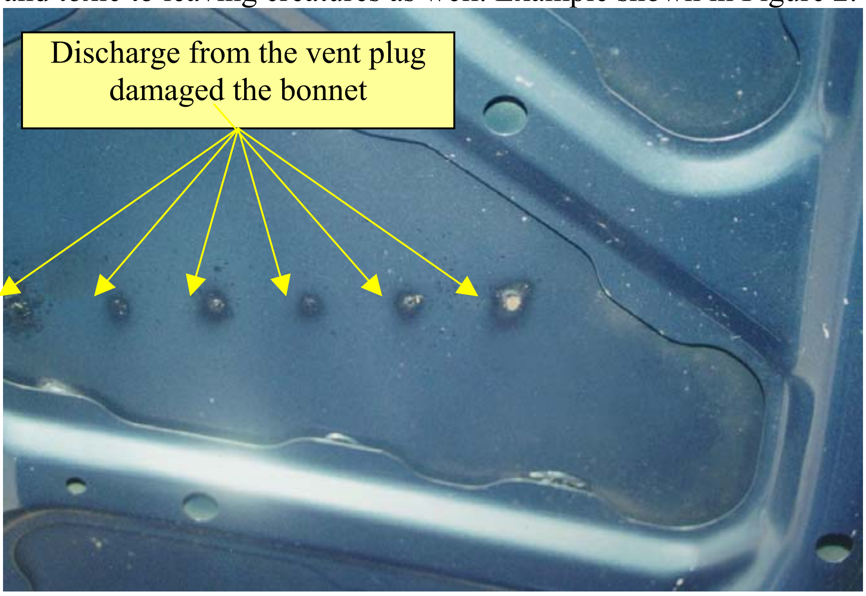

Figure 2: Battery acid causes permanent damage to metal parts

On top of that, it will also cause the vehicle damage and involved high repair cost for welding, painting and etc. Subsequently, it will jeopardize the value of the vehicle in the near futures. The acid on the battery has actually discharge through ventilation hole and cause discharge to the battery compartments. In some serious condition, it may also penetrate to the chassis of the vehicle cause the chassis to collapse, due to the highly corrosive acid as shown in Figure 3.

Figure 3: Battery acid discharged to the vehicle chassis

Another issue is the symptoms of the swelling maintenance free battery due to the high temperature especially in the tropical countries as shown in Figure 4.

Figure 4 : Typical problems on maintenance free battery (bulged/ swollen)

Safety is another paramount concern and enormous strength that has been built to prevent corrosion in the battery compartment and lead poisoning, when leakage of the battery acid. Lead acid discharged to the environment cause pollution, which is harmful to human as well as other leaving things. However, up till today, those batteries which claimed to be sealed tight batteries usually, face the problems of unreliable and very short serviceable life span when the batteries are used in the tropical climate. The basic reason of the batteries failure is due to 'Thermo Runaway'.

Sealed tropicalized lead acid battery by applying the application of the gas separation membrane is proposed to solve the century old electrolyte decomposition and corrosive problems in the lead acid batteries industries as well as environmental issues.

The membrane with gas separation technology, which acts as a selective barrier, will allow certain gas component permeate faster than the other one in a gas mixture. Therefore, by using the similar concept, the membrane technology will used to retain electrolyte level by controlling or minimised the rate of vaporised electrolyte disposal into the atmosphere. Hydrogen and oxygen gas found during charging and discharging will be permeating through membrane as to balance the pressure build- up inside the cells, or else explosion might be occurred. The membrane pore size, thickness and effective area need to be sized for the battery with different Ampere Hour (AH) capacities.

Principles of Lead Acid a battery is a device that converts the chemical energy contained in its active materials directly into electrical energy by means of and oxidation-reduction electrochemical reaction. This type of reaction involves the transfer of electrons from one material to others. In non-electrochemical reactions, this transfer of electrons occurs directly and only heat is involved. In a battery, the negative electrode or anode is the components capable of giving up electrons, being oxidized during the reaction. It is separated from the oxidizing material, which is the positive electrode or cathode, the components capable of accepting electrons. The transfer of electrons takes place in the external electric circuit is connected to the two materials as a load. Transfer of charge is completed within the electrolyte by movement of ions.

The electromotive force (EMF) producing active materials of lead-acid storage battery is lead dioxide (PbO_2), lead (Pb), and sulphuric acid (H_2SO $2H \rightleftarrows + SO_4$). Chemically, the elementary lead has the valence zero and lead dioxide is a quadrivalent lead and, in sulphuric acid, both of them is unstable but rich in chemical energy and, if permitted to gain or give up electrons, will start reacting with sulphuric acid to change into a stable bivalent lead (lead sulphate).

A concise description of the theory of electromotive force (EMF) production by the lead-acid storage battery is that this chemical change of lead dioxide and lead into lead sulphate gives rise to a flow of electrons (electric current) in the external circuit, which may also be expressed as a conversion of chemical energy to electric energy[2].

Characteristic of a Lead Acid Battery during Discharging. The operation of cell during discharge is shown schematically in Figure 5. When the cell is connected to an external load, electrons flow from the anode, which is oxidized, through the external load to the cathode, where the electrons are accepted and the cathode material is reduced. The electric circuit is completed in the electrolyte by the flow of anions (negative ions) and cations (positive ions) to the anode, respectively. The discharge reaction can be written as below, whereas lead (Pb) as the anode material and lead oxide (PbO_2) as the cathode material. Finally, we get the overall reaction for discharge as in Equation (1.3):

At negative electrode: anodic reaction (oxidation, loss of electrons)

$$Pb \rightarrow Pb^{2+} + 2e \quad (1.1)$$

At positive electrode: cathodic reaction (reduction, gain of electrons)

$$PbO_2 + 4H^+ + 2e \rightarrow Pb^{2+} + 2H_2O \quad (1.2)$$

Overall reaction: (discharge)

$$Pb + PbO_2 + 2H2SO_4 \rightarrow 2PbSO_4 + 2H2SO_4 \quad (1.3)$$

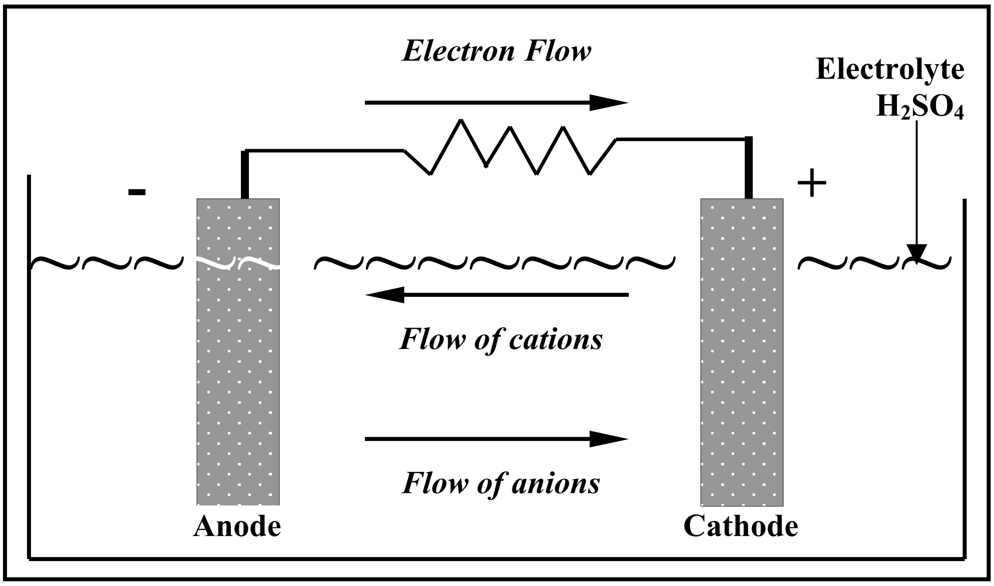

Figure 5 : Cell operations during charging state

Characteristic of a Lead Acid Battery during Charging. During recharging of a secondary battery such as lead acid battery, the current flow is reversed and oxidation takes place at the positive electrode and reduction at the negative electrode, as shown in Figure 6. As the anode is, by definition, the electrode at which oxidation occurs and the cathode the one where reduction takes place, the positive electrode is now the anode and the negative is the cathode.

At negative electrode: cathodic reaction (reduction, gain of electrons)

$$Pb^{2+} + 2e \rightarrow Pb \quad (1.4)$$

At positive electrode: anodic reaction (oxidation, loss of electrons)

$$Pb^{2+} + 2H_2O \rightarrow PbO_2 + 4H^+ + 2e \quad (1.5)$$

Overall reaction: (charge/ recharge)

$$2PbSO_4 + 2H_2O \rightarrow Pb + PbO_2 + 2H2SO_4 \quad (1.6)$$

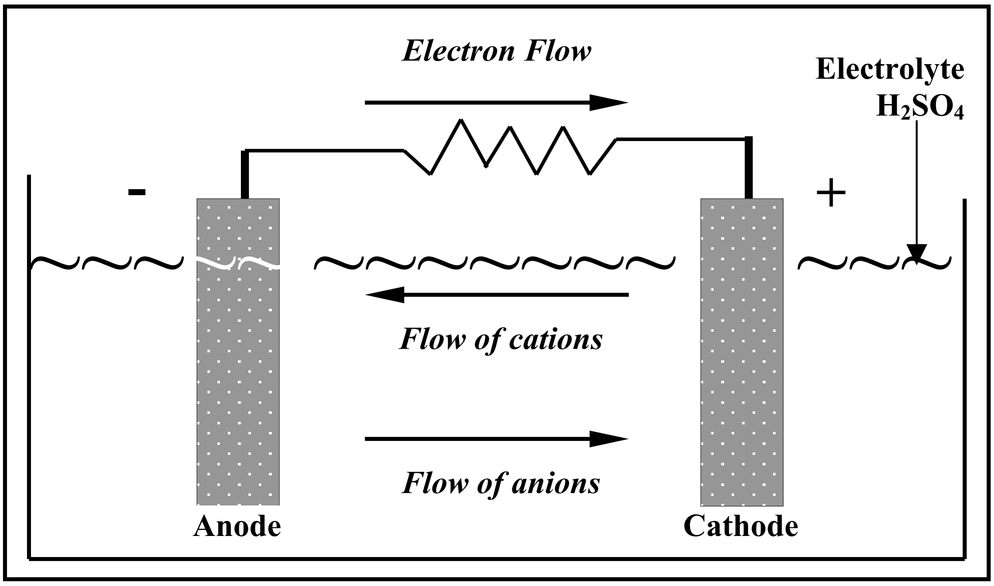

Figure 6: Cell operations during charging

Nevertheless, there is another side of reaction that occurs during charging of lead acid battery instead of the primary reduction-oxidation (redox) reaction as mentioned above. During the final stage of charging, whenever the cell nearly approaches full charge, or when most of the lead sulphate ($PbSO_4$) has been converted into lead (Pb) and lead oxide (PbO_2), the cell voltage will become greater than the gassing voltage. At this point and onwards, overcharge reactions occurs, and as a resultant, hydrogen and oxygen is produced at anode and cathode. The reaction, which is more known as the gassing reaction, is shown as the following equations[2]:

At negative electrode

$$2H^{+} \quad + \quad 2e \quad \rightarrow \quad H_2 \qquad (1.7)$$

At positive electrode

$$H_2O \quad \rightarrow \quad \frac{1}{2}O_2 \quad + \quad 2H^{+} \quad + \quad 2e \qquad (1.8)$$

Overall reaction

$$H_2O \quad \rightarrow \quad H_2 \quad + \quad \frac{1}{2}O_2 \qquad (1.9)$$

This gassing reaction, however, is said by battery researchers to be the main factor to the electrolyte decomposition in lead acid batteries[2].

Influences of Electrolyte Decomposition in Lead Acid Batteries[4]. It is aforementioned that the main reason for the decomposition of electrolyte from the lead acid battery is caused by electrolysis due to high rate charging and discharging. However, the electrolysis phenomenon occurs normally whenever the battery is charged. The basic electrode processes in the positive and negative electrodes involve a dissolution-precipitation mechanism and not some sort of solid-state ion transport or film formation mechanism. As the sulphuric acid in the electrolyte is consumed during discharge, water is produced, the electrolyte can be considered an “active material” and in certain battery, designs can be the limiting material[4].

As the cell approaches full charge and the majority of the lead sulphate ($PbSO_4$)has been converted to laed (Pb) and lead oxide (PbO_2), the cell voltage on charge becomes greater than the gassing voltage and the overcharge reactions begin, resulting in the production of hydrogen and oxygen (gassing) and the resultant loss of electrolyte.

Current Method of Controlling Electrolyte Losses in Lead Acid Batteries[4]. Engineers have tried all sorts of means to control decomposition in lead acid batteries from all the battery industries around the world. With years of research, some of the methods which are found believe will work under certain circumstances and conditions. Listed below are the four most common types of methods to control electrolyte losses in lead acid batteries.

(i) Replace Antimony Grid with Calcium Grid for Suppression of Gaseous[4].
(ii) Additional Volume of Electrolyte in Lead Acid Batteries[9].
(iii) Valve Regulated Lead Acid Batteries[6].
(iv) Substitute Liquid Electrolyte with Gel Electrolyte[5].

Many workers have been studying on how to suppress the hydrogen evolution and oxygen reduction in lead acid batteries. This is because the main factor that causes gassing reaction to be occurring in lead acid battery. During the production of lead acid battery, additives were introduced into lead paste to make it more durable and strong, as lead is a very soft substance. Normally, substances like antimony and calcium were used added in lead paste to the earlier mentioned objective. However, these substances cause marked influence on gassing reaction to occur.

2. Experiments

The research consists of various kinds of evaluation that must be done in sequence in order to obtain the best results which will represent the entire research parameters. Basically, the whole research methodology is divided into three segments as below: (Refer to Figure 7)

(i) Battery technology
(ii) Membrane technology
(iii) Battery with membrane technology

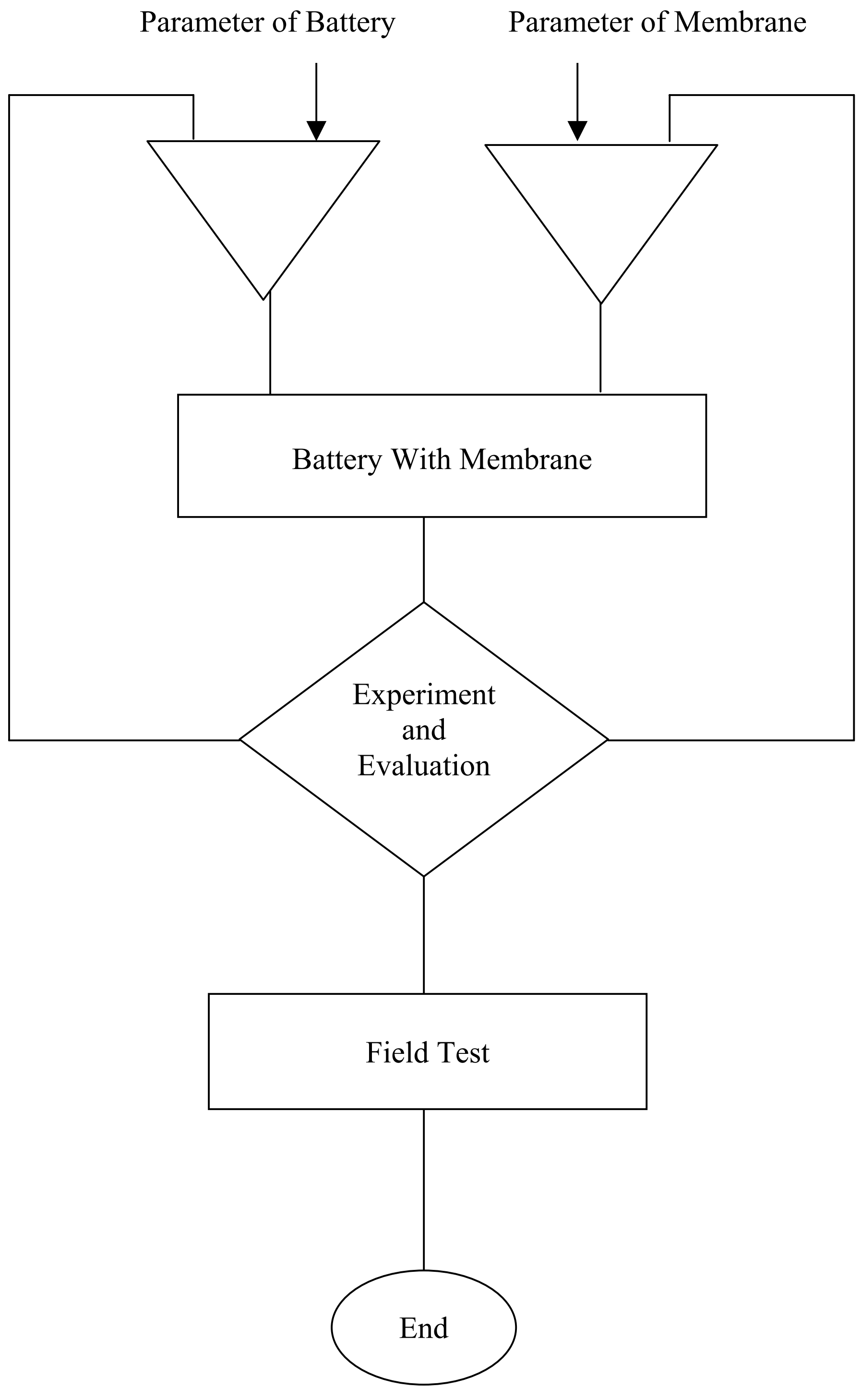

Figure 7: Overall flow chart of research methodology

2.1. Lead acid battery and equipments

Firstly, we need to gather sufficient and informative data in order to differentiate and identified the root of the cause or the problems before conduct any experiment. The accuracy of the results on the research is highly depends on the key problems, Hence, the accuracy of the data input must be very precise in order to obtain reliable scrupulous results. Equipments used have been calibrated within specification by the master calibrator before used.

This experiment is based on the current automobile batteries and the related supporting systems such as, electrical generator/alternator of the vehicle operating under the tropical conditions. Of course, the condition could be like the weather, humilities, temperatures and etc. An experiments will be carried out by using Malaysia's one of the most popular car. This research is based on the Proton Wira 1.5L, the equipments has been installed inside the vehicle while driving; the purpose is to obtain the best accurate parameter from the vehicle. The location of sensors has been installed and set up inside the engine compartment for picking-up data while driving.

2.1.1. Description of sensors and equipments:

(i) Voltage Probe:- Connected to battery positive terminal to measure the charging voltage.
(ii) Temperature Probes:- Insert to the centre cell, in between plates. It is to measure the accumulation of the temperature between the plates.
(iii) Temperature Probe (Battery surface):- Attached to the battery container surface as to measure the fast transient temperature movements.
(iv) Temperature Probes (Air Temperature) :- Installed at the back of the radiator which is also in between the radiator and the battery.
(v) Temperature Probe (Engine Temperature):- Installed close to the exhaust manifold, as to monitor the heat radiated form the exhaust manifold.
(vi) Intelligent Digital Multimeter:- This is to indicate real time voltage at the battery terminals. This indicated the rate of changes and loads by the Alternator with running engine.
(vii) Data Recorder:-. All signals from the Temperature Probe (Thermocouple) are feed to the Data Recorder for data collection.
(viii) Interfaced Cable:- Wire connection from the sensors to the equipments.
(ix) Digital Oscilloscope:- Used to measure the ripple waveform generated by charging alternator.
The recorded results the above is tabulated in Table 1

Table 1
The results obtained for the case study.

Descriptions	Result	Remarks
Voltage probe (1)	10.32 – 14.83V	Changing Voltages. Rate of charge
Temperature Probe (2) inserted into cell	82 ºC – 84 ºC	Temperature rise slowly but sustain on for long period even is stopped after 2 hours.
Temperature Probe (3) Battery casing surface	65 ºC – 115 ºC	Response rapidly to the engine R.P.M and road speed.
Temperature Probe (4) engine compartment	80 ºC – 135 ºC	Temperature is low when vehicle is moving at high speed.
Temperature Probe (5) engine manifold	280 ºC – 385 ºC	Temperature variation according engine speed
Charging Ripple	37 – 45 Vpp. 860 Hz – 9 KHz	Both Vpp and *f* varies according to the engine speed without heavily load.

2.2. Materials

Actually, there are various types of membrane and also applications found in the literature reviews[7,8]. However, Gas Separation Membrane has been found to be most suitable technology and subsequently was chosen to be the only application in this research for tropicalized lead acid batteries.

Before casting of a flat sheer membrane can be made, selection of materials and solvent/non-solvent has to be finalised and prepared in the first place. In general, a casting solution will consist of a polymer, a non-solvent and a solvent.

As for this research application, polysulfone has been selected as base polymer for all the casting solution made. As for the solvent, N, N-Dimethylacetamide (DMAC), Dimethyformamide (DMF) and Tetrahydrofuran (THF) have been selected due to the strong interaction with Polymer and miscibility with water. Meanwhile the non-solvent used in this experiment is Aliphatic Alcohol, Ethanol. It is used as a non-solvent additive for Polysulfone(PSF)

Polysulfone (Udel Bisphenol A Polysulfone Udel P1700) from Amono Performance Products was being used as one of the membrane materials because of the easy commercial availability, and a favourable selectivity permeability characteristics, good mechanical strength, thermal property and acid durable.

And other chemicals such as N, N-Dimethylactanride (DMAC), Dimethylformamide (DMF) Ethanol and Tetrahydrofuran (THF) were laboratory grade from Aldrich Co.

2.3. Electrolyte decomposition test with ac component

The gases generated during charging and the life span of battery is also heavily depending on the percentage Ripple present during charging. Ripple is an unwanted AC. (Alternating Current) component which carried by the DC. (Direct Current) supply to the battery under charged. The purity of DC supplies sources, which cause vibration of both positive and negative electrodes, thus shedding of lead pasting on the grid. As result of the high ripple presents, the evolution of gasses is much higher as compare to the one with pure DC charging process. The evolution of gasses is due to agitation effect which is the vibration from the line frequency[12]. The result is tabulated in Table 2a and 2b.

Table 2.(a)
Weigh losses per hour of charge at 25 ºc

Charging Current	Charger With Ripple Of Line Frequency 50Hz			Charger With Pure DC Source		
Amp	Weight Of Electrodes Losses (gm)/Hour					
	Before	After	% loss	Before	After	% loss
1	860	857	0.35	860	860	0
3	857	843	0.60	860	860	0
6	843	812	3.70	860	858	0.23
9	812	767	15.5	858	853	0.58
12	767	603	21.4	853	846	0.82

Table 2.(b)
Electrolyte losses per hour of charge at 48 ^{o}c

Charging Current	Charger With Ripple Of Line Frequency 50Hz			Charger With Pure DC Source		
Amp	Weight Of Electrolyte (gm)/Hour					
	Before	After	% loss	Before	After	% loss
1	550	543	0.36	550	549	0.18
3	548	532	2.91	549	547	0.36
6	532	500	6.01	547	542	0.94
9	500	447	10.6	542	533	1.67
12	447	396	17.5	533	527	3.00

3. Results and discussion

3.1. Membrane enhanced lead acid battery simulation test

After completion of the preliminary test, it has known that the formulation of 12.5 wt % of Polysulfone (PSF) and 87.5 wt % of N-N-dimethylformamide (DMF) were the more suitable membrane that is going to be tested on conditional simulation test. The actual condition test consists of a Pulse Width Modulated (PWM) charger and a hot bath simulator. The battery was immersed in the hot bath simulator with temperature set at 85 ^{o}C and it was charged by a Pulse Width Modulated (PWM) charger. The test condition and parameter is shown in Table 3

In the initial of the first stage, experiment has been established with the suitable membrane formulation and this membrane is applied onto the battery for further evaluation. After various types of membrane are fabricated, those membranes are then installed with membrane specimens jig fixture as to find out whether they are capable of maintaining electrolyte level with minimum pressure built-up. In order to put the membrane to the real condition test, the membrane is attached onto the battery vent hole and then the battery is charged by the Pulse Width Modulation (PWM) charger that have similar charging pattern on the alternator of the vehicle. In addition to that, the battery is dipped into the hot water bath simulator with water level just before the vent plugs. At the sometime, it has to set the temperature controller Set Value (SV) to 85^{o}C. This is to simulate the condition under the bonnet that combines the high charging rate and scorching hot temperature inside the battery compartment. As refer to the earlier research results, those are the two main parameters that contribute to the decomposition of electrolyte.

Table 3
Parameter used for the lead acid battery simulator test

1	Battery type JIS	NS60, made by Watta
2	Battery weight Dry	8.50 kg
3	Battery weight Wet	12.80 kg
4	Electrolyte volume	≈ 3.2 L or 0.53 L per cell
5	Elect. Level min. max	≈ 80ml between min. and max.
6	Charging voltage DC	14.40 V means. 14.60 V max.
7	Charging ripple	≈ 40 Vpp
8	Ripple Frequency	≈180 Hz – 2 KHz
9	Charging temperature	85°C max
10	Electrolyte to weight ratio	1 liter ≈ 1.34 kg

The flow rate of the gaseous permeating from the battery during the charging without applying any membrane increased continuously. In Figure 8 shows that, electrolyte is continuously decomposed without any application of membrane. Meanwhile the flow rate of the gaseous permeating from the battery during the charge with the application of the membrane is slowly rising at the early stage of charging and the rate of increasing saturated as time elapses. Hence, this indicates that wherever charging goes along the way, the permeation of the gaseous will continuously increase and decomposition of the electrolyte will take place, if the lead acid battery is not enhanced with membrane technology.

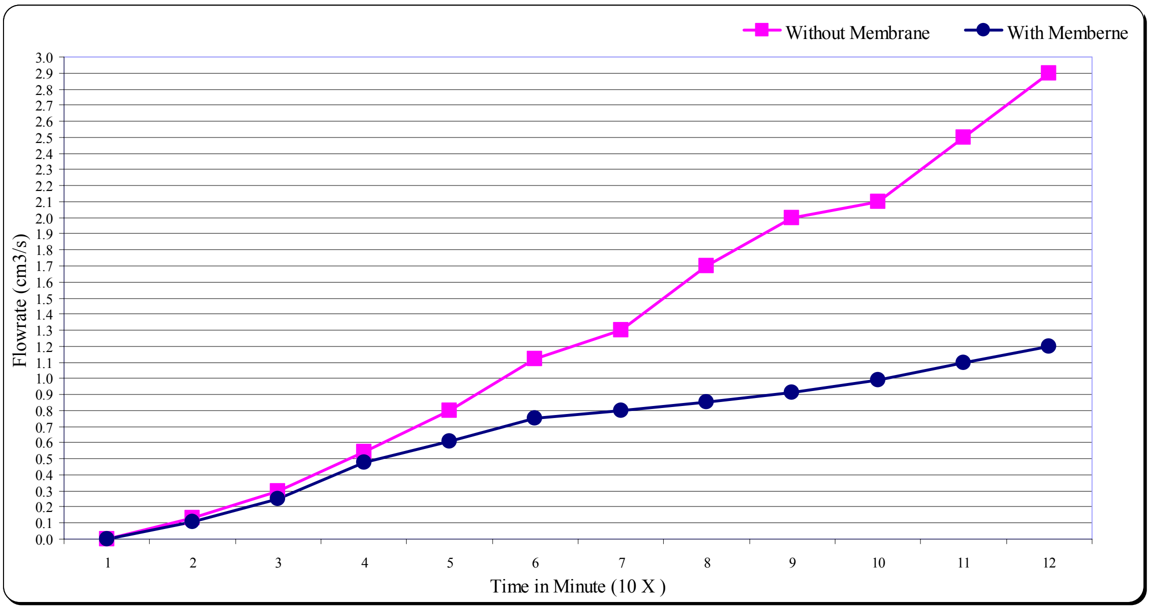

Figure 8: Electrolyte losses during stimulated battery charging. (comparing with and without membrane)

3.2. Electrolyte losses during stimulated battery charging

The rate of the electrolyte losses from the battery with the membrane was much slower than the rate of the battery that does not have the membrane. As based on acuminated data, the average rate of the electrolyte losses over a period of 168 hours is approximately 0.0095 Grams per hour, while the rate of the electrolyte losses from the battery that does not use any membrane is approximately 0.024 Grams per hour. In another words, when a membrane is applied to the battery's vent hole, it has proven that it is able to retain the electrolytes from getting vented to the atmosphere. As the result, this means that the membrane is able to be used as an electrolyte retaining device in the lead acid battery.

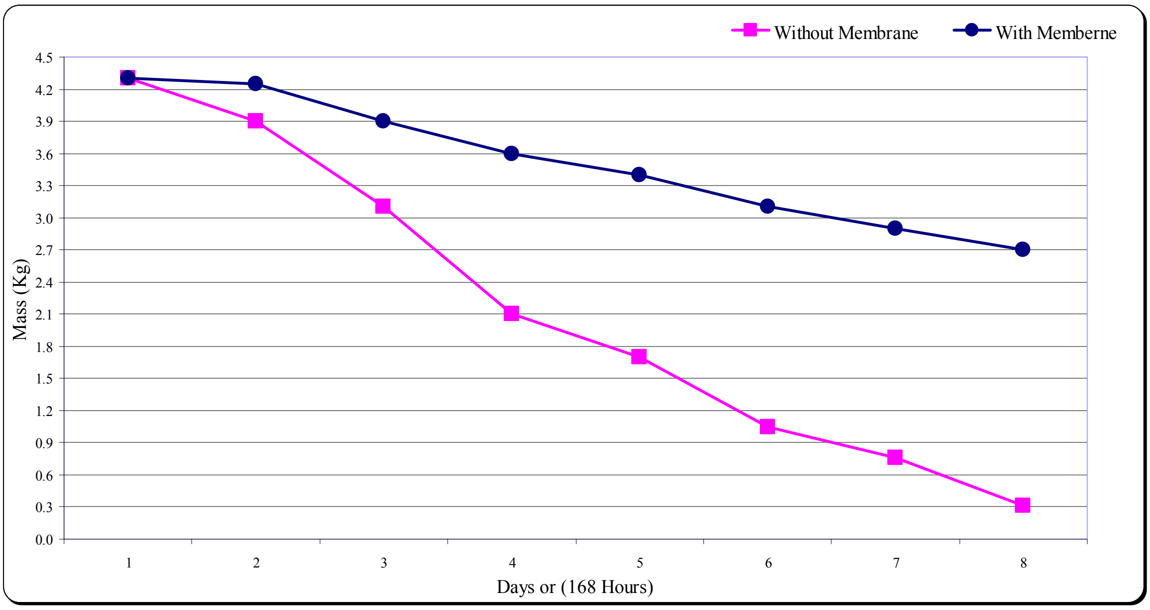

Figure 9: The Weight of electrolyte losses in the battery while charging. (comparing with membrane and without membrane).

From data shown in Figure 9. we summarized , the electrolyte weight losses in the battery without membrane is approximately 0.24 Grams per hour or 0.576 Grams per day Therefore, the electrolyte will fall below the minimum require level of the battery after 7 days of usage, base on the initial weight of the electrolyte at 4.3Kg. Take into the considerations that both of Anode and Cathode Plates were dry when it is new, which mean both plates will consume some percentage of electrolytes. In order the cells to be activated, both plates' need to be 100% immersed into electrolyte. It is also proof that the battery enhanced the membrane is capable to control electrolyte losses.

4. Conclusion

With referring to the results compiled from the laboratory and field experiments, it is illustrated the asymmetric gas separation membrane, shown the success in the development of the application on the tropicalzed lead acid battery in maintaining the electrolyte level for the whole serviceable life span. However, there is no doubting that the results are extremely encouraging. It seems to confirm that the membrane works.

We are confident that, when the membrane is full developed battery life will be extended and explosions will be eradicated.

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