

**THE EFFECTS OF CELL POROSITY AND THICKNESS UPON THE
PERFORMANCE OF LITHIUM-ION BATTERY**

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in partial fulfillment of the requirements for the degree of
Bachelor of Mechatronics Engineering with Honours**


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DECLARATION

I declare that this thesis entitled "THE EFFECTS OF CELL POROSITY AND THICKNESS UPON THE PERFORMANCE OF LITHIUM-ION BATTERY is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

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APPROVAL

I hereby declare that I have checked this report entitled "THE EFFECTS OF CELL POROSITY AND THICKNESS UPON THE PERFORMANCE OF LITHIUM-ION BATTERY" and in my opinion, this thesis it complies the partial fulfillment for awarding the award of the degree of Bachelor of Mechatronics Engineering with Honours

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19 JUN 2019

DEDICATIONS

To my beloved mother and father

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ABSTRACT

This study focuses on the effect of cell porosity and thickness upon the performance of lithium ion batteries (LIBs). Nowadays, the demand of high efficiency of Lithium-ion batteries is growing as the result of exponential growth of electronic devices among consumers and in industrial sectors. The fact of the exposure of LIBs to open air could cause an explosion and small batteries capacities become its massive drawback. Hence, the purpose of this study are to produce and verify the numerical solutions of LIBs modelling for LiFePO₄ half-cell cathode in order to analyse the effect of cell porosity (ε_v), radius of particles (a_0), and thickness (L) upon the performance of lithium-ion batteries by observing the discharge curves. One of the components to achieve high performance LIBs is by optimizing the geometry of the cell. A mathematical models of LIBs based on drift diffusion model takes into account the electrolyte equation and lithium transport equation in the electrode particles which incorporated the geometry of cell microstructure to the coefficients in a macroscopic model. The multi-scale model is solved numerically using Finite Difference Method (FDM), and the simulated discharge curve for 0.5C and 1C discharge current have been verified with experimental data. The effect of geometry variation on the discharge curve are examined by varying three parameters namely porosity (ε_v), size of particles (a_0) and cell thickness (L) while other parameters are remained same. As results, the constructed simulation shows that the higher volume fraction (porosity content) produces large discharge curves while decreasing the size of electrode particles increase the capacity of the cell. Meanwhile, less electrode thickness is most likely to give better battery performance as compared to wide thickness of electrodes.

ABSTRAK

Kajian ini bertujuan untuk mengkaji kesan keliangan sel and ketebalan terhadap prestasi bateri lithium-ion (LIBs). Pada masa kini, permintaan bateri lithium-ion yang berprestasi tinggi semakin meningkat kerana hasil peningkatan eksponen peranti elektronik dalam kalangan pengguna dan sektor perindustrian. Permasalahan bateri meletup apabila didedahkan kepada udara and mempunyai kapasiti kecil menjadi permasalahan terbesar kepada LIBs. Oleh itu, tujuan kajian ii dijalankan adalah untuk menghasilkan and membuktikan ketepatan penyelesaian berangka model LIBs untuk sel positif LiFePO₄ demi mengkaji kesan keliangan cell (ϵ_v), saiz zarah (a_0) and ketebalan (L) terhadap prestasi bateri lithium-ion dengan memerhatikan lengkungan penyahcas. Salah satu cara untuk mencapai LIB berprestasi tinggi adalah dengan mengoptimumkan reka bentuk geometri sel. Model matematik LIBs berdasarkan model penyebaran hanyut mengambil kira persamaan elektrolit dan persamaan pengangkutan litium dalam zarah elektrod yang memasukkan reka bentuk geometri mikrostruktur sel kepada pekali dalam model makroskopik. Model multi-skala diselesaikan secara berangka dengan menggunakan Kaedah Perbezaan Hingga (FDM), dan lengkung nyahcas untuk 0.5C dan 1C arus nyahcas dijana dan telah disahkan dengan menggunakan data eksperimen. Kesan variasi reka bentuk geometri pada lengkung nyahcas diperhatikan dengan mengubah tiga parameter iaitu porositi (ϵ_v), saiz zarah (a_0) dan ketebalan sel (L) manakala tiada perubahan berlaku kepada parameter lain. Kajian mendapati bahawa, simulasi dibina menunjukkan bahawa keliangan yang lebih tinggi (kandungan porositi) menghasilkan lengkung nyahcas yang besar dan kapasiti sel meningkat apabila saiz zarah dikurangkan. Sementara itu, ketebalan elektrod yang rendah berkemungkinan untuk menjana prestasi bateri yang lebih baik berbanding dengan ketebalan elektroda yang tinggi.

TABLE OF CONTENTS

	PAGE
DECLARATION	
APPROVAL	
DEDICATIONS	
ACKNOWLEDGEMENTS	ii
ABSTRACT	iii
ABSTRAK	iv
TABLE OF CONTENTS	v
LIST OF TABLES	vii
LIST OF FIGURES	viii
LIST OF SYMBOLS AND ABBREVIATIONS	x
LIST OF APPENDICES	xi
CHAPTER 1 INTRODUCTION	1
1.1 Overview	1
1.2 Motivation	1
1.3 Problem Statement	4
1.4 Hypothesis	5
1.5 Objectives	5
1.6 Scope and Limitation	5
1.7 Thesis Outline	6
CHAPTER 2 LITERATURE REVIEW	7
2.1 Overview	7
2.2 Lithium-ion Batteries	7
2.3 Electrochemical Mechanism of Lithium-ion Batteries	9
2.3.1 Charge Separation on SEI Layer	9
2.4 Types of Lithium-ion Batteries	11
2.5 Active Materials in Lithium-ion Batteries (LIBs)	14
2.5.1 Electrodes	14
2.5.2 Electrolytes	15
2.6 Half-cell	17
2.7 Modelling of Lithium-ion Batteries	18
2.7.1 Modelling for Half-cell Cathode	21
2.8 Numerical Method	21
2.9 Thickness and Porosities in Lithium-ion Batteries	23
2.10 Summary	24

CHAPTER 3	METHODOLOGY	25
3.1	Overview	25
3.2	Project Flowchart	25
3.3	Analysis Description	27
	3.3.1 Finite Difference Approximation Method (FDM)	28
3.4	Verification of Numerical Simulation	29
3.5	Numerical Simulation	31
CHAPTER 4	RESULTS AND DISCUSSIONS	32
4.1	Overview	32
4.2	Comparison between Full-cell and Half-cell	33
	4.2.1 Error Analysis	34
4.3	Numerical Analysis	36
	4.3.1 Effect of Porosity Distribution upon the Performance of LIBs	36
	4.3.2 Effect of Particles Size upon the Performance of LIBs	39
	4.3.3 Effect of Thickness upon the Performance of LIBs	40
4.4	Summary	42
CHAPTER 5	CONCLUSION AND RECOMMENDATIONS	43
5.1	Conclusion	43
5.2	Future Works	43
	REFERENCES	44
	APPENDICES	46

LIST OF TABLES

Table 2.1	The lithium diffusivity in electrodes active materials [10]	14
Table 2.2	Electrical conductivity in active electrode materials [10]	15
Table 2.3	Physical properties of organic carbonate solvent [17]	15
Table 2.4	Ion conductivity ($S\ cm^{-1}$) of salt solution [17]	16
Table 2.5	List of symbols used in modelling of LIBs	19
Table 3.1	List of Discharge Current for Different C-rate	30
Table 3.2	The physical parameter for LiFePO ₄ electrode and LIPF ₆ electrolyte [3]	30
Table 3.3	The transport properties of electrode and electrolyte [3]	30
Table 3.4	The Value of Varying Parameters in Each Case	31
Table 4.1	List of residual values of numerical simulation and experimental data comparison for 0.5C and 1C.	35

LIST OF FIGURES

Figure 1.1	The Classification of Batteries	2
Figure 1.2	Market Segmentation by Geography in 2018 [3]	3
Figure 1.3	Market Segmentation by Application in 2018 [3]	3
Figure 2.1	Schematic Diagram of the Structure of LIBs [11]	8
Figure 2.2	Structure of the electric double layer	10
Figure 2.3	The spinel structure in positive-electrode of LiMn_2O_4	11
Figure 2.4	The scale for the overall performances of LIBs	13
Figure 2.5	The half-cell structure proposed by Newman	17
Figure 3.1	Flowchart for Project Methodology	26
Figure 3.2	Discharge curves of different C-rate obtained from Yu <i>et al</i> [4]	29
Figure 4.1	The comparison of numerical simulation (solid lines) of LiFePO_4 half-cell cathode with full-cell experimental data at different discharge rates.	33
Figure 4.2	The residual values curves of numerical simulation and experimental data comparison for 0.5C and 1C.	34
Figure 4.3	The microstructure of particles arrangement in electrode in 2D	36
Figure 4.4	The discharge curve of the 1C discharge rate when volume fraction, ε_v is varies into ε_v (green), $\varepsilon_{v(\max)}$ (red) and $1/2\varepsilon_{v(\max)}$ (blue).	37
Figure 4.5	The discharge curve of the 1C cell when particles sizes are varies into a_0 (green), $1/2a_0$ (red) and $2a_0$ (blue).	39

- Figure 4.6 The discharge curve of the 1C cell when cell thickness, L is varies into L (green), $1/2L$ (red) and $2L$ (blue). 41
- Figure 4.7 The concentration gradient of electrolyte solution across electrode at $t=100s$. 41

LIST OF SYMBOLS AND ABBREVIATIONS

LIBs	-	Lithium-ion batteries
SOD	-	State of Discharge
C-rate	-	Current rate
LiFePO ₄	-	Lithium Iron Phosphate
LiPF ₆	-	Lithium Hexafluorophosphate
LiCoO ₂	-	Lithium Cobalt Oxides
LiMn ₂ O ₄	-	Lithium Manganese Oxides
Li ₄ Ti ₅ O ₁₂	-	Lithium Titanate
NMC	-	Lithium Nickel Manganese Cobalt Oxides
NCA	-	Lithium Nickel Cobalt Aluminium Oxides
SEI	-	Solid-electrolyte interface
EC	-	Ethyl carbonate
DMC	-	Dimethyl carbonate
PDE	-	Partial Differential Equations
FDM	-	Finite Difference Approximation Method
ODE	-	Ordinary differential equation
RMSE	-	Root mean square error
Li^+	-	Lithium-ion
e^-	-	Electron
N^-	-	Neutral ion
c_s^*	-	Lithium-ion concentration in solid
c^*	-	Lithium-ion concentration in electrolyte
ϕ_s^*	-	Solid potential of cell
ϕ^*	-	Electrolyte potential of cell
U_{eq}^*	-	The equilibrium cell potential
j_s^*	-	Current density in electrode
j^*	-	Current density in electrolyte
x^*, L	-	Cell thickness
t^0	-	Transference number
R	-	Universal gas constant
F	-	Faraday constant
b_{et}	-	BET surface area
D	-	Diffusion coefficient
G^*	-	Reaction rate inside cell
κ	-	Electrical conductivity
σ_s	-	Ionic conductivity of Lithium-ion at solid phase
r, a_0	-	Radius of electrode particle
η^*	-	Over-potential of cell
ε_v	-	Volume fraction of electrolyte (Cell porosity)

LIST OF APPENDICES

APPENDIX A	GANTT CHART	46
APPENDIX B	POTENTIAL COMPARISON BETWEEN NUMERICAL AND EXPERIMENTAL DATA AT 0.5C	47
APPENDIX C	POTENTIAL COMPARISON BETWEEN NUMERICAL AND EXPERIMENTAL DATA AT 1C	48

CHAPTER 1

INTRODUCTION

1.1 Overview

This chapter presents the introduction, motivation, problems statement, hypothesis, objective, scope and limitation of the proposed project.

1.2 Motivation

Lithium-Ion Batteries are classified into two categories; primary and secondary batteries. Both are used as power source but they have different electrical recharge ability. The primary battery is designed for one-time uses. This means these kinds of batteries are not rechargeable once they reach their maximum C-rate due to irreversible electrochemical reaction inside the LIBs. Normally found in portable electronic devices such as toys and memory backup; the long lifespan, lightweight, high energy density and easy to use became the advantages of primary batteries. Besides, this type of batteries also required less maintenance compared to secondary.

While secondary battery is rechargeable as they can reverse the electrochemical reaction inside the battery. When recharging, current must be applied in opposite direction of the discharge current. High discharge rate, power density and efficient low temperature performance becomes the advantages of using the secondary cells. However, secondary batteries have poor energy density compared to primary. Based on Bajagain[1], the application of secondary batteries falls under two categories. The first category is using the secondary batteries as energy storage devices, where the battery supplies energy following demands from load while being charged through primary sources such as solar panel or electric grid. While the second categories are the batteries are discharged as primary battery

and can be fully recharged again[1]. There are four classes of chemicals usually found in secondary batteries which is lead acid, Nickel-metal-hydride (NiMH), Nickel-cadmium (NiCd) and Lithium-ion Batteries. However, this work only takes account on Lithium-ion Batteries or known as LIBs.

The usages of Lithium ion batteries (LIBs) as products power source are quite popular among electronic device producers. In last three decades, LIBs have gained world attention due to their characteristics such as high energy density, rate of charge and discharge, reliability and design flexibility[2]. As most electronic and electric device operates in long duration, they need an energy storage that offered high energy density and capacity such as LIBs. Despite having large energy capacity, the LIBs are considered affordable in term of prices due to unlimited source and low maintenance. There are two possible users of LIBs which is consumers and industrial manufacturer. Both have different perspective in choosing LIBs specification. While consumer seeking for high capability, industrial application require battery system that have a good loading capabilities, long life and provide safe and dependent service.

Besides, self-discharge rate also considered as important trait in choosing a wise battery type as this could be major problem to cell performance. Cell with lower self-discharge rate has the longer the lifetime. Although self-discharge is not considered as manufacturing defect, poor fabrication or handling could increase the rate of self-discharge in cell. The amount of self-discharge varies with battery type. Normally, the cell with above 30% self-discharge has to be disposed immediately. Satisfying the characteristic, the lower rate of cell self-discharge becomes one of the advantages in LIBs. Moreover, design flexibility can be done to allow the LIBs to maximize its performance in longevity, specific power and specific energy.

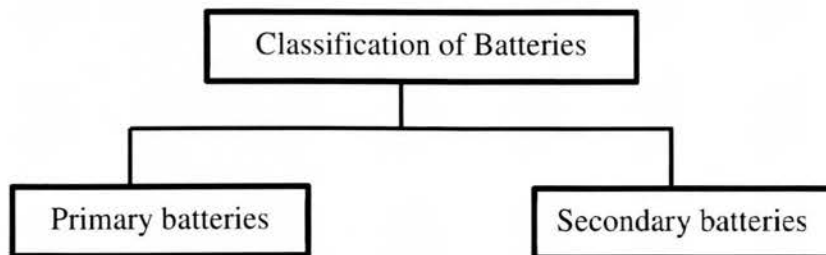


Figure 1.1 The Classification of Batteries

According to P&S Market Research[3], the exponential growth of electronic and electrical devices among consumers or industrial sectors helps to increase the demands of Lithium-ion Batteries (LIBs) in marketplace as in 2018, the market of LIBs reached \$33,720.8 million. Since the industrialization of portable devices grows rapidly, higher efficiency of LIBs are needed for these applications. In addition, LiFePO₄ batteries are recorded to have the largest market share due to their high application usage in automotive application such as electric vehicle, uninterruptible power supply (UPS) system and stationary energy storage system.

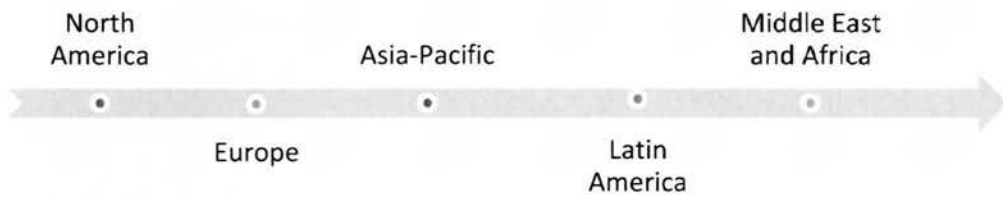


Figure 1.2 Market Segmentation by Geography in 2018 [3]

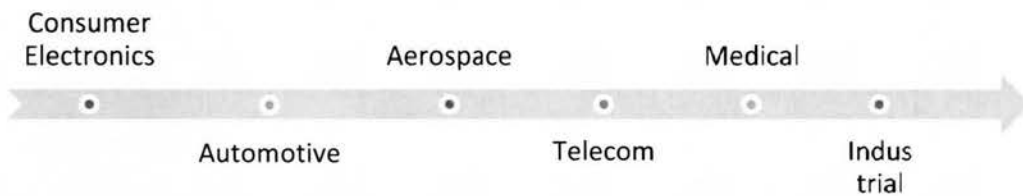


Figure 1.3 Market Segmentation by Application in 2018 [3]

Figure above listed the market segmentation in geography and application of LIBs global market for period of 2014-2018. The batteries properties advancement, prices and demand become the key trend for LIBs market fluctuation. Additionally, changes of lifestyles preferences and attraction toward smart electronic devices especially for newer generation are the reason for high market demand for LIBs and while studying for the mechanism of LIBs, most research such as Yu *et al*[4], Yang[5] and Du *et al*[6] covers on improving the properties of active materials to enhance LIBs performance. However, little research done on varying the geometry of the cell. Thus in this research, the geometry and characteristic of the battery are discussed in order to improve the performance of the LIBs.

1.3 Problem Statement

The fact of the exposure of LIBs to open air could cause an explosion becomes its massive drawback. Besides, it also requires regular recharging as the commercialized LIBs capacities are usually small. In contrast, the presence of “super battery” can be a paradigm shift to the electronic and electrical devices manufacturer. The term “super battery” is defined to the ideal battery characteristics such as having large specific energy, specific power and capacity but lower in resistance. Besides, it is expected by consumer to have a gadget that able to be fully charged instantly without being overheating as much as having low discharge rate. However, the “super battery” is not yet to be found until this day and the existed excellent-performance batteries are expensive.

In order to understand and improving the drawback of LIBs, the geometry of the cell must be studied. Before any analysis is carried out, there is a concern regarding on how to increase the LIBs performance by varying the geometry aspect of the cell. The characteristic that expected to be manipulated are the porosity distribution (ε_v), radius of particles (a_0), and thickness (L) of cells. However, the manual way to measure the self-discharge for LIBs is by measuring the decrement of the cell open circuit voltage over time [7]. As LIBs have small discharge rate, this ways is considered very time consuming as huge amount of time needed to measure discharge rate of samples. In addition, the cell fabrications for samples are considered expensive in term of preparation and costing. This concern also can be applied to any other analytical experiment in determining LIBs performance.

Other way to measure the effectiveness of varying porosity distribution (ε_v), radius of particles (a_0), and thickness (L) to the LIBs performance is by using a mathematical modelling. However, the usage of this method also has its own limitation due to high complexities and time consuming to solve the partial derivation equations (PDE). In addition, the results of this method have to be verified with a real-time analysis or experiments before it can be fully accepted. In short, the model complexities, time varies and method disadvantages are considered as major challenges in this work.

1.4 Hypothesis

For electrodes materials, cells with high porosity, ε_v content and smaller particle size, a_0 are expected to produce better performance compared to cells with lower porosity content and small surface area. While for the thickness (L) of electrodes, smaller thickness value is most likely to give better battery performance as compared to wide thickness of electrodes.

1.5 Objectives

The purposes of this study are:

1. To produce numerical solutions of Lithium-ion Batteries (LIBs) modelling for LiFePO₄ half-cell cathode using Finite Difference Approximation Method (FDM).
2. To verify the numerical simulation by comparing the discharge curves with full-cell experimental data.
3. To analyse the effect of cell porosity (ε_v), radius of particles (a_0), and thickness (L) upon the performance of lithium-ion batteries by observing the discharge curves.

1.6 Scope and Limitation

This study is based on the mathematical model described by Newman[8], [9] and homogenized to 1D by Richardson. The LIBs is analysed in half-cell. For the half-cell cathode, Lithium Iron phosphate (LiFePO₄) is used as positive electrode and lithium foil as its reference electrodes. For the electrolyte, standard electrolyte material such as Lithium Hexafluorophosphate (LiPF₆) is chosen for both half-cell anode and half-cell cathode.

This mathematical model is based on drift diffusion model which relates the electrolyte equations and lithium transport equations with the microstructure geometry of electrode particles. Using MATLAB software, the numerical model are solved using Finite Difference Approximation Method (FDM) by referring

Rahifa[10] as guide. The simulated discharged curve is then verified using experimental data obtained from Yu *et al*[4].

This analysis are expected to focus on finding the effect of geometry variation of three different electrode parameters; porosity (ϵ_v), radius of particles (a_0), and thickness (L) as these parameters are believe influences the cells diffusion rate. Based on these parameters variation, the performances of LIBs are studied by observing the discharge curve.

1.7 Thesis Outline

This research consists of five chapters. The first chapter explains the purpose of this study in analysing the effect of cell porosity (ϵ_v), radius of particles (a_0), and cell thickness (L) upon the performance of LIBs.

Chapter 2 covers the theoretical background of Lithium-ion battery (LIB) and reviews on related studies done by other researchers. This chapter also explains the details of model circuitry and geometry design in LIBs.

Chapter 3 discussed the methodology of the analysis. Flowchart and Gantt chart are constructed to show the estimated progress timeline. In addition, the methods of analysis are explained to give a clearer view of this work.

While Chapter 4 discussed the expected result may obtained in this analysis through the experimentations done in Chapter 3. In addition, battery modelling for electrode and electrolyte in various conditions are explained in details to give deep understanding to readers.

And lastly, Chapter 5 concludes all the finding of this work. Recommendations of future works are also described in this chapter.

CHAPTER 2

LITERATURE REVIEW

2.1 Overview

This chapter provides details explanation of Lithium-Ion battery. The mechanism and working principal are explained. The electrochemical battery modelling and methods for determining cells self-discharge value proposed in previous works are introduced. Review on previous works related to the effect of cell porosity (ε_v), radius of particles (a_0), and cell thickness (L) upon the performance of Lithium-Ion batteries (LIBs) are studied and presented on this chapter.

2.2 Lithium-ion Batteries

Lithium-Ion Batteries or LIBs is one of the power supply for electronic devices that converting chemical energy into electrical energy. The consistency in supplying DC voltages while discharging process makes battery such as Lithium-Ion Battery a suitable replacement of portable power generator. Besides, features exhibit by the cell such as low cost, large capacity, high durability, charge and discharge rate, intrinsic stability, design flexibility and outstanding environmental compatibility makes LIB a popular choice among electronics manufacturer.

Based on Figure 2.1, the basic components in LIB are anode, cathode, separator and electrolyte. Made from metal elements, electrodes act a bridge that forges the charge to travel on. There are two types of electrodes polarity which is cathode; a positively charged electrode that often referred as working electrode and anode; a negatively charged electrode that often referred as counter electrode.

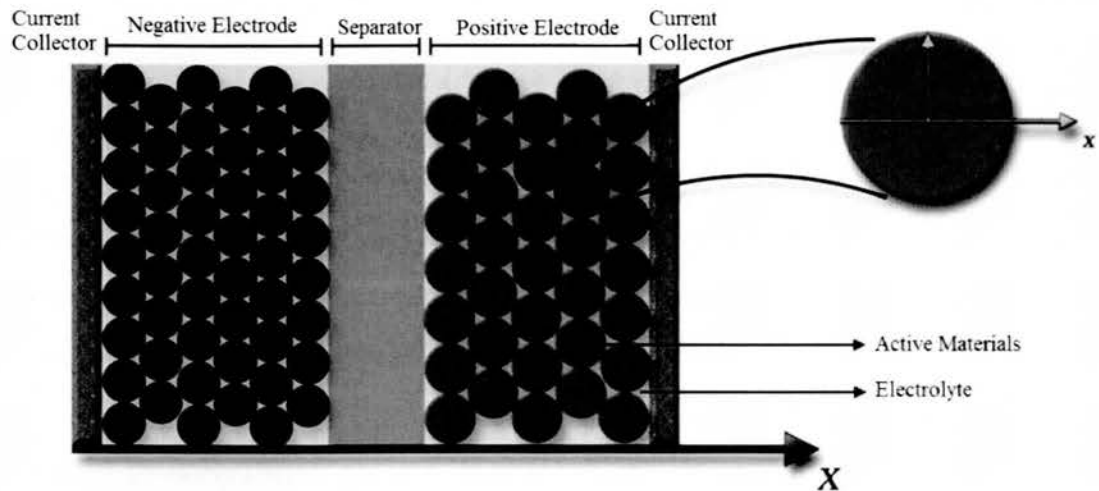
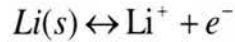


Figure 2.1 Schematic Diagram of the Structure of LIBs [11]

While, separator is a porous polymeric film that lies between two electrodes that acts as insulator and prevents the direct electrical contact so there is no occurrence of short circuit in cells and electrolyte consist of “ionically conductive” organic solvent that provide pathway to Li^+ . The porous structure of separator allows the passage in electrolyte solution.

The current collector plays important role in transferring the electrons charge to the outer circuit efficiently. The common material used for current collectors are copper and aluminium. Porous current collectors are important to increase surface area, reduce the significant mass of the battery and improving electronic conductivity of cells [1]. Cells cannot complete its cycle if the structure is incomplete. Thus, these components are considered important as the cell cannot be functioning if the basic structure is incomplete.

2.3 Electrochemical Mechanism of Lithium-ion Batteries



During discharging, the solid particles, $Li(s)$ in anode diffuse and formed Lithium-ion, Li^+ and electron, e^- on the solid-electrolyte interface (SEI) layer. The electron is released to electrode particle and follows its pathway to the current collector. While, Li^+ is released to the electrolyte solution and diffuse through the separator in order to reach cathode. The ion transportation of both electrons and Li^+ is called de-intercalation process. In contrast, the intercalation process takes place when the free Li^+ in electrolyte solution is attracted to the SEI layer of electrode particle in cathode and takes an electron with it. In short, it can be concluded that the neutral ion, N^- remain in the electrolyte while Li^+ carry charges throughout the route in LIBs [10]. However, the process is reversed during charging process due to external forces caused by electrical source. The external forces must supplied higher voltage than voltage produce by cell so the current can move in reverse direction and cause the ion transportation to moves from cathode to anode; where the intercalation process happens.

2.3.1 Charge Separation on SEI Layer

Solid-electrolyte interface (SEI) is a thin layer exists around solid particle in electrode and its formation protects the electrode against solvent decomposition at large voltage. As stated in Pinson *et al* [12], this layer growth on the surface of active materials is the result irreversible electrochemical decomposition of the electrolyte. The structure of SEI on active materials is known as electric double layer and the simplest model is introduced as Helmholtz model (refer Figure 2.2).

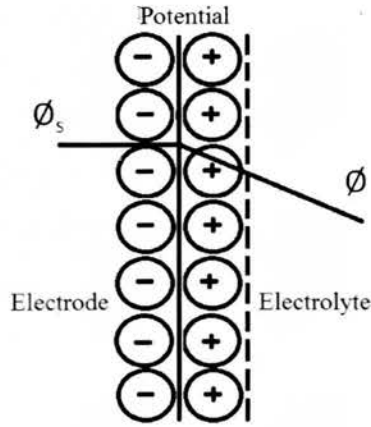


Figure 2.2 Structure of the electric double layer

On this structure, the electric drop linearly from electrode potential, ϕ_s^* to the electrolyte potential, ϕ^* [10]. As the charge neutrality moves freely in electrolyte solution as bulk, the excess Li^+ are expected to attached on the anode surface; leaving the negative ion, N^- in electrolyte. It can be concluded that, LIBs would face continuous loss of Li^+ as ion transportation are impossible without presence of SEI layer. The over-potential in electric double layer structure is described as:

$$\eta = \phi^* - \phi_s^* + U_{eq} \quad (2-1)$$

The symbol ϕ^* stand for electrolyte potential while ϕ_s^* is the solid potential in electrode. In addition, U_{eq} is the equilibrium potential of the electrode material or called electrode open-circuit potential. Next, the continuous losses of Li-ion in diffusions are known as capacity fade. For LIBs, the average capacity fade is between 0.025 – 0.048% per cycle. Based on research done by Pinson *et al*[12], the most common source of irreversible capacity fade in manufactured LIBs occurs on SEI layer, which typically forms at the negative electrode during recharging.