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The effect of electrolyte parameter variation upon the performance of lithium iron phosphate ($LiFePO_4$)

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ABSTRACT

This study focuses on the effect of electrolyte parameter variation upon the performance of lithium iron phosphate ($LiFePO_4$) due to high demand of the electronic devices among consumers and industrial sectors that require efficicent $LiFePO_4$ batteries. Hence, it is essential to improve its performance by optimizing the electrolyte parameter of the cell. The mathematical models of lithium-ion batteries based on the drift-diffusion model consists of the electrolyte and lithium transport equations in the electrode particles which incorporated the geometry of cell microstructure to the coefficients in the macroscopic model was solved numerically using the method of lines (MOL) technique. The effect of electrolyte parameter variation on the discharge curve of $LiFePO_4$ using lithium hexa-fluoro-phosphate ($LiPF_6$) as the base electrolyte is examined by varying the electrolyte parameters: initial concentration of lithium ions (c_0), the effective diffusivity of lithium ions (D) and the effective ionic conductivity (κ). The simulations show that higher diffusivity of lithium ions produces large discharge curve and higher ionic conductivity increase the mobility of the ion in the cell. Meanwhile, higher initial concentration of lithium ions produced high battery performance compared to lower initial concentration of lithium resulting more discharge time before electrolyte depletion occurs.

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1. INTRODUCTION

Lithium-ion battery (LIB) uses an intercalated lithium compound as the material of positive electrode and graphite as the negative electrode. The demand for lithium-ion batteries (LIBs) has increased tremendously since year 2005 as LIBs have conquered most markets for rechargeable batteries and have been used in many applications of portable electronics such as mobile devices, laptops, and electrical appliances [1]. This gained a lot of attention from researchers around the world in looking at the development of lithium batteries as it is a wide potential in automotive industries which investigate their use in electric vehicles; markets for all of which are expected to expand exponentially in the future [2], [3].

Hence, improvement in the performance of lithium-ion batteries is essential. In lithium-ion batteries, the choice of the electrolyte solution is an important consideration for both safety and performance. While studying for the mechanism of LIBs, most research focus on enhancing the properties of electrodes to improve LIBs performance and infrequent studies have been done on varying the electrolyte parameter of the cell [4]. Several studies have suggested that factors of limiting the discharge of LIBs are electrolyte depletion and fully discharge of electrode particles [5]-[7].

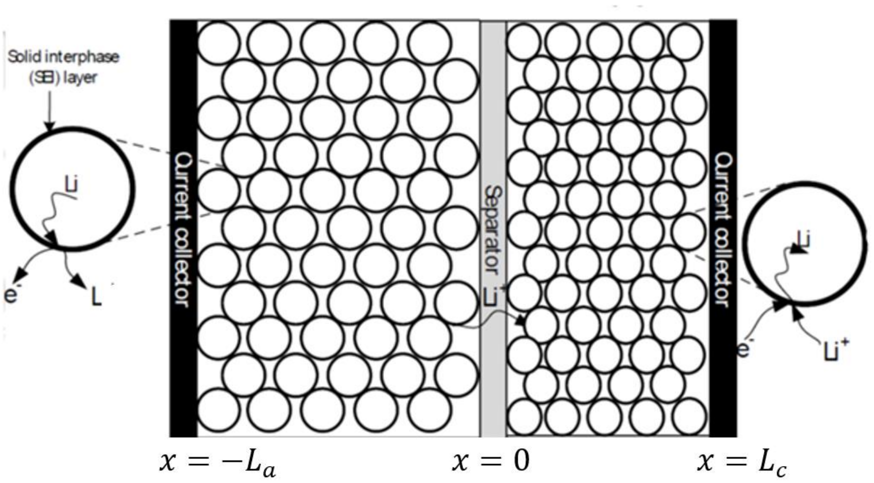

Figure 1. A schematic diagram of lithium-ion battery during discharge which sandwiches between an anode $(-L_a < x < 0)$, a separator and a cathode $(0 < x < L_c)$ [7]

A lithium-ion battery which is abbreviated as LIB is a type of a rechargeable battery. It is one of the power supplies for electronic devices that converted from chemical energy to electrical energy. They are commonly used for portable devices such as cellular phones, digital camera and computer and are also popular for use in electric vehicle, military and aerospace applications because of their high performance with high capacity, low self-discharge, reliability and high output voltage [1]. Based on Figure 1, lithium-ion battery consists of five major components which are anode, cathode, separator, current collector and electrolyte, where r is the radial coordinate and x is the thickness of the cell.

Anode and cathode are known as electrode which are made from metal element and acts as a bridge that makes the charge to travel. During discharge, the anode which consists of negative electrode particles is where the loss of electrons occur (oxidation) while cathode which contains of positive electrode particles is where the gains of electrons occur (reduction). The separator that lies between the two electrodes prevents direct electrical contract between the two electrodes to avoid shirt circuit while allowing the passage of electrolyte. At the outer boundaries of electrode, there are current collector or also known as charge collectors which transfer electron charge through the external circuit. Electrolyte, which transports ion between cathode and anode, is the last major component in the lithium-ion battery.

The discharge of the battery starts when the de-intercalation mechanism occurs at electrode particle surface in anode to release a lithium ion and an electron into the electrolyte. The electron travels from anode to cathode current collectors via the external circuit while the lithium ion diffuses and advects through the electrolyte across the separator and gets into cathode. The intercalation process occurs at the electrode partical surface in cathode under a similar reaction and diffuses into the particle in radial axis. We note that to test the behaviour of a specific electrode material, a half-cell experiment is performed where the test material as working electrode and the other electrode is set up as reference electrode [4]. Thus, in this paper, we focus on the half-cell of $LiFePO_4$ as the working electrode while Li-foil (anode) acts as reference electrode where we denote $x = 0$ is the separator and $x = L_c$ is the thickness of the cathode. The electrodes and separator are bathed in $LiPF_6$ electrolyte which is a common electrolyte of LIBs [8], [9].

The battery modelling is a multiscale system that describes the conservation of ions (Li^+) in the electrolyte, charge reaction at the electrode surface $(Li^+ + e^- \rightleftharpoons Li_s)$ and transport of lithium (Li_s) within the electrode particles [6], [7]. Richardson *et al.* [10] used homogenization techniques to derive the lithium-ion battery (LIB) model in dilute electrolyte which incorporates the macroscopic system (the thickness of cell) and the microscopic system (the size of electrode particles). The mechanisms in LIB for a particular behaviour can be solved using suitable model as discussed in [11]. However, the electrochemical modelling has the most accuracy in anticipating the behavior of LIB but with disadvantage of it is computationally expensive [12].

Understanding the dynamic processes that occur during charge and discharge of the battery is essential to improve the power density and cycle life of the battery. Study of the charge/discharge processes is made more complicated by the multiscales model, scaling from the macroscopic scale of battery thickness to the microscopic scale of the radius of the electrode particle [13], [14]. The reaction kinetic for the deintercalation/intercalation of lithium at the electrode particle surface may be limited by diffusion of Li^+ in the electrolyte, diffusion of Li_s in the electrode particles or by the electrical resistivity of the electrolyte or electrodes [15]-[17]. Thus, it is important to investigate the relative importance of these processes.

Several studies have been conducted to investigate the charge transport behaviour of $LiPeFO_4$ which is one of the highest demanding LIBs. The $LiFePO_4$ materials have the characteristic of a flat discharge curve that shows no variation of voltage until it is fully discharged [18]. The aging behavior in a graphite/$LiFePO_4$ cell was stimulated. It is found that the aging is dominantly manifests itself in terms of capacity loss rather than in terms of impedance increase [19]-[21]. Rahman *et al.* [22] used particle swarm optimization to segregate various battery parameter that exhibits vast variation under utmost solution to the model which gives a practical instrument. In other investigation, a lithium phase transport in electrode particle of $LiFePO_4$ cathode was conducted which resulted to be the premier limiting factor of capacity loss in $LiFePO_4$ [23].

In this research, the electrolyte parameters and characteristics of the electrolytes are discussed to improve the performance of the $LiFePO_4$. Here, we vary the diffusion coefficient of lithium ion and the initial concentration of lithium ion in the electrolyte while other parameters are fixed.

2. LITHIUM BATTERY MODEL

The Newman electrolyte model is being used in this research and is under approximation of charge neutrality in the bulk electrolyte except in layer near the electrolyte/electrode particle interface named Debye layer. The model applied the Stefan-Maxwell multicomponent diffusion equation, and the electric potential follows Poisson's equation for the derivation. The details of the derivation can be found in [7], [15], [16]. The model accounts for the over-potential at the electrode surface described in (1), the η gives the change in the electrochemical potential of lithium ions between the electrolyte and the electrode. In (2) describes the drift diffusion of lithium ions in electrolyte where c and ϕ denote as concentration of Li^+ and electrolyte potential, respectively. In (3) is the conservation of charge and is given by the gradient of the average current density (j) in the electrolyte. The reaction rate, G of the intercalation at the electrode surface is given by the Butler Volmer equation (see (4)). A lithium-ion electrolyte can be fully characterized, in terms of the electrolyte model (2) to (5), by experimental measurement of transference number (t_+) and ionic diffusivity and conductivity as function of concentration of lithium ion ($D(c), \kappa(c)$). In (5) and (6) denote the transport of lithium in the electrode particle in radial axis where c_s and j_s are the concentration of lithium and electrode current density, respectively. The boundary conditions and initial conditions for the governing (1) to (6) are found in (7) to (10). The equilibrium potential of $LiFePO_4$ as a function of concentration of lithium $U_{eq}(c_s)$ is given by (11).

$$\eta = \phi - \phi_s + U_{eq} \tag{1}$$

$$\varepsilon_v \frac{\partial c}{\partial t} = \frac{\partial}{\partial x}\left(D(c)\beta \frac{\partial c}{dx}\right) - \frac{\partial t_+^0}{\partial x}\frac{j}{F} + (1 - t_+)b_{et}G \tag{2}$$

$$\frac{\partial j}{dx} = Fb_{et}G, where:$$

$$j = -\beta\kappa(c)\left(\frac{\partial \emptyset}{\partial x} - 2\frac{RT}{F}(1 - t_+)\frac{\partial \log(c)}{\partial x}\right) \tag{3}$$

$$G = i_0(c, c_s)\left(\exp\left(-\frac{F\eta}{2RT}\right) - \exp\left(\frac{F\eta}{2RT}\right)\right)$$

$$i_0(c, c_s) = k_0(c)(c_s|_{r=a})^{1/2}(c_{max} - c_s|_{r=a})^{1/2} \tag{4}$$

$$\frac{\partial c_s}{\partial t} = \frac{D_s}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial^2 c_s}{\partial r}\right) \tag{5}$$

$$\frac{\partial j_s}{\partial x} = -Fb_{et}(x)G, where:$$

$$j_s = -\sigma_s \frac{\partial \phi_s}{\partial x} \tag{6}$$

$$-D_s \frac{\partial c_s}{\partial r^*}|_{r=a} = G(c, c_s|_{r=a}, \phi - \phi_s) \tag{7}$$

$$c|_{x=0} = c_0, \left.\frac{dc}{dx}\right|_{x=L_c} = 0 \tag{8}$$

$$\mathrm{\phi}|_{\mathrm{x=0}} = 0, \mathrm{j}|_{\mathrm{x=L_c}} = 0 \tag{9}$$

$$\mathrm{j_s}|_{\mathrm{x=0}} = 0\ \mathrm{j_s}|_{\mathrm{x=L_c}} = -\frac{\mathrm{I}}{\mathrm{A}} \tag{10}$$

$$\mathrm{U_{eq}(c_s)} = 3.114559 + 4.438792 \arctan(-71.7352\mathrm{c_s} + 70.85337) - 4.240252 \arctan(-68.5605\mathrm{c_s} + 67.730082) \tag{11}$$

The descriptions of all the parameters are listed in Table 1. In modelling the half-cell $LiFePO_4$ battery, lithium hexa-fluoro-phosphate, $LiPF_6$ is used as the electrolyte, since it has high conductivity and stability [24]. The electrolyte parameter is determined from the literature review, either from the journal or articles. The three parameters are found which are the initial concentration of lithium ions (c_0), the effective diffusivity of lithium ions (D) and the effective ionic conductivity (κ). The diffusivity (in (2)) and conductivity (in (3)) of the electrolyte as concentration dependant are fitted to the equations [7].

$$D(c) = D_0 e^{-0.0003017c} m^2 s^{-1} \tag{12}$$

$$\kappa(c) = \kappa_0 c(5.2069 - 2.14 \times 10^{-3} c + 2.3440 \times 10^{-7} c^2 AV^{-1} m^{-1} \tag{13}$$

Table 1. List of symbols in battery model

Parameter	Description	Parameter	Description
$D(c)\ (m^2 s^{-1})$	Diffusivity of lithium ions	$\phi(JC^{-1})$	Electric potential in the electrolyte
ε_v	Particle volume fraction	$x\ (m)$	Length
$c\ (mol\, m^{-3})$	Concentration of lithium ions	r	The radial coordinate of electrode particles
t_+	Transference number of lithium	$j\ (Cs^{-1} m^{-2})$	Current density
$t\ (s)$	Time	$T\ (K)$	Temperature
$F\ (C\, mol^{-1})$	Faraday's constant	$A\ (m^2)$	Cross-sectional area of electrode
$R\ (J\, mol^{-1} K^{-1})$	Universal gas constant	$G(\, mol\, m^{-2} s^{-1})$	Reaction kinetic at the electrolyte/electrode particle interface
$\phi_s(JC^{-1})$	The potential of electrode particles	$i_o(c, c_s)(Am^{-2})$	Exchange current density
$b_{et}(m^{-1})$	BET surface area	$I({C_s}^{-1})$	The total charge flowing across the surfaces of the electrode particles
$c_s(mol\, m^{-1})$	Intercalated lithium concentration in electrode particles	a	The radius of electrode particles
$U_{eq}(c_s)(JC^{-1})$	Equilibrium potentials	L_c	The thickness of cathode
D_s	The diffusitivity of lithium in electrode particles		

3. NUMERICAL SCHEMES

Method of lines (MOL) technique is used to solve (1) to (13). This technique transforms the partial differential equations (PDEs) in (1) to (6) into a set of ordinary differential equations (ODEs) in time derivative by descritising the spatial derivatives using centered difference approximation of finite difference method. The ODEs are then be solved with respect to time using the built-in solver 'ode15s' in MATLAB. In (4), the reaction rate depends on the lithium ion concentration (c) and intercalated lithium concentration (c_s) at the electrode surface. The numerical scheme is written in assumption that there is a spherical particle (diffusion of intercalated Li_s within the radius of spherical particle, $r = a$) at each grid points of x (the thickness of cell) and the particles touch each other as shown in Figure 2. Therefore, the size of the numerical scheme (the differentiation matrix) depends on the number of n grid points of x, and the number of representative spherical particles which is descretised radially by m grid points of r. For example, if we take $n = 100$ grid points of x and $m = 100$ grid points of r for each spherical particle, we generate 10300×10300 entries of the differentiation matrix. The resulting differentiation matrix is a large system, therefore a Jacobian matrix is applied to improve the efficiency of the solver [7].

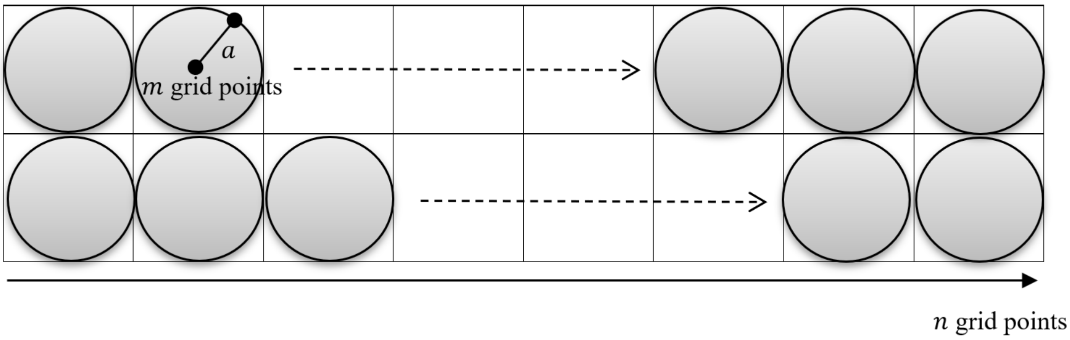

Figure 2. The grid points of numerical scheme

4. RESULTS AND DISCUSSION

The results obtained explains the charge transport behavior inside LiFePO4 electrode material during discharge process. The transport process usually determines the performance of a Lithium-ion battery. The species transport includes the lithium-ion concentration in the electrolyte and the intercalated lithium in the electrode particle. The state of discharge (SOD) is the ratio of cell capacity that has been discharged to the cell maximum capacity denotes by $0 \leq y \leq 1$. The parameter values used in the model are listed in Table 2.

The simulation of the model has been verified in Ranom [6], [7]. The numerical simulation is compared with the $LiFePO_4$ experimental data Yu et. al [25]. It is proven that the simulation of the model shows good comparison to the experimental data. The electrolyte diffusion is taken to be $10^{-10} m^2 s^{-1}$ based on the value calculated by Valøen and Reimers [24] and the size of the particle is less than 1 micrometer which reflects the tortuosity and porosity of the composite electrode structure. On the other hand, the intercalated lithium diffusion coefficient in the $LiFePO_4$ particle used the value $10^{-18} m^2 s^{-1}$ which shows rapid diffusion within the electrode particles.

Figure 3 depicts the discharge curves for different discharge rates. The drop of voltage as discharge rate increases occurs because of the capacity loss due to charge transport limitation in the electrolyte and electrode particles. The charge transport limitation in electrolyte happens when electrolyte depletion region appears in the Li^+ concentration profiles. The depletion region avoids the intercalation process, thus hampering the discharge (see Figure 4). The depletion region in electrolyte prevents the active material in electrode particles being discharged at the particle surface. In electrode particles, the process of intercalation at the electrode particle surface increases very fast (because of rapid diffusion in the electrode particles) leads to fully discharge of particle (full of intercalated lithium) in the region near to the separator, thus slowing down the discharge in the underlying region as shown in Figure 5.

Table 2. The parameters value [6], [7]

Parameter	Value
Electrolyte parameter	
Diffusivity of lithium ions $D_0 (m^2 s^{-1})$	5.253×10^{-10}
Volume fraction ϵ_v	0.4764
Initial salt concentration $c_0 (molm^{-3})$	1000
Transference Number t_+	0.38
Ionic conductivity $\kappa_0 (AVm^{-1})$	10^{-4}
Electrode parameters	
Radius of particle $a(m)$	5.2×10^{-8}
Diffusion coefficient in the solid $D_s (m^2 s^{-1})$	6×10^{-18}
Exchange current density $i_0 (Am^{-2})$	5.4×10^{-5}
BET surface area $bet\ (m^{-1})$	5×10^7
Maximum concentration in the solid $C_{s,max} (molm^{-3})$	20950
Other parameters	
Electrode thickness $L\ (m)$	6.25×10^{-5}
Faraday constant $F\ (Cmol^{-1})$	96487
Universal gas constant $R\ (Jmol^{-1}K^{-1})$	8.3144
Temperature $T\ (K)$	298
Electrode area $A\ (m^2)$	10^{-4}

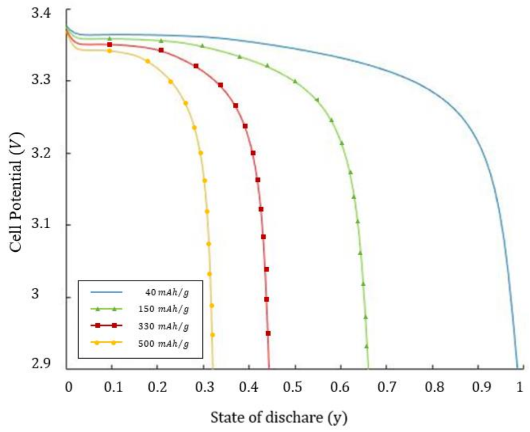

Figure 3. Discharge curves for different discharge rates

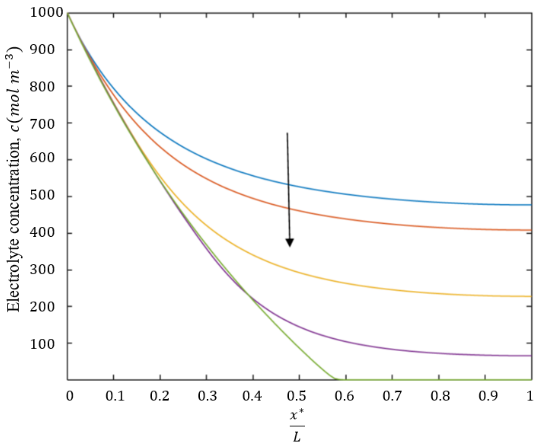

Figure 4. The concentration of Lithium-ion, Li^+ in the electrolye accross the dimensionless thickness of electrode at 150 mAh/g discharge rate. The arrow shows the direction of time increases at t^* =344s, 688s, 1032s, 1376s, 1720s

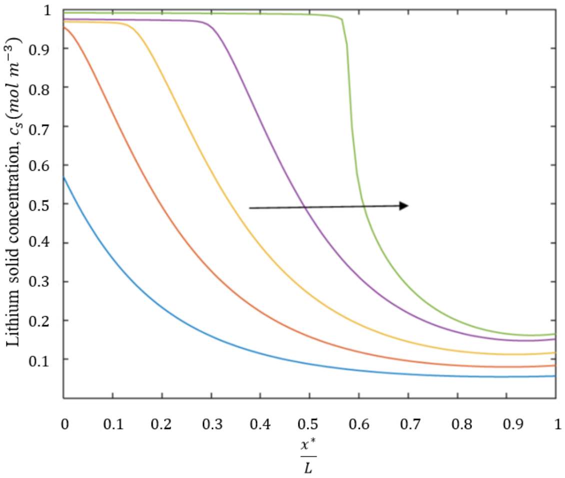

Figure 5. The dimensionless concentration of lithium in the solid, Li_s at the electrode particle surfaces as a function of dimensionless position in the electrode at 150 mAh/g discharge rate. The arrow shows the direction of time increases at $t^* =$ 344s, 688s, 1032s, 1376s, 1720s

4.1. Effect of initial concentration upon the performance of LIBs

Figure 6 presents the discharge curves of LiFePO$_4$ electrode when the initial concentration is varied to $800\,molm^{-3}, 1000\,molm^{-3}$ and $1200\,molm^{-3}$. The slightly lower initial concentration $c_0 = 800\,molm^{-3}$ shows poor discharge curve which end approximately at 0.56 state of discharge due to the quick development of electrolyte depletion region. As we increase the initial concentration, $c_0 = 1200\,mol\,m^{-3}$ the cell performance increases by 14% which ends approximately at 0.77 SOD. The increment of cell performance is expected as it delays the time for the electrolyte to become depleted.

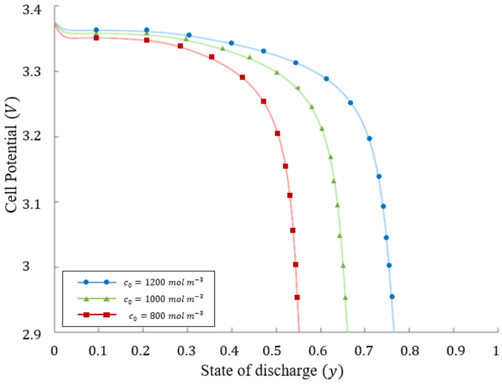

Figure 6. The discharge curve at 150 mAh/g discharge rate when initial concentration, c_0 is varied

4.2. Effect of diffusivity of lithium ions upon the performance of LIBs

Figure 7 shows the effect of varying the Lithium-ion diffusitivity upon the LiFePO$_4$ cell performance. Here, we varied the diffusitivity value from the $D_0 = 3 \times 10^{-11} m^2 s^{-1}$ by half ($D_0 = 1.5 \times 10^{-11} m^2 s^{-1}$) and double the value ($D_0 = 6 \times 10^{-11} m^2 s^{-1}$). It can be seen that the cell with the highest diffusivity $D_0 = 1.5 \times 10^{-11} m^2 s^{-1}$ shows the best performance with the highest capacity compared to lower diffusivity. Furthermore, the capacity loss occurs at the early discharge especially at low Lithium-ion diffusivity. The effect of the Lithium-ion diffusitivity is quite significant as depicted in Figure 7. As the diffusitivity increases, the performance of LiPeFO$_4$ electrode material increases by almost 30%.

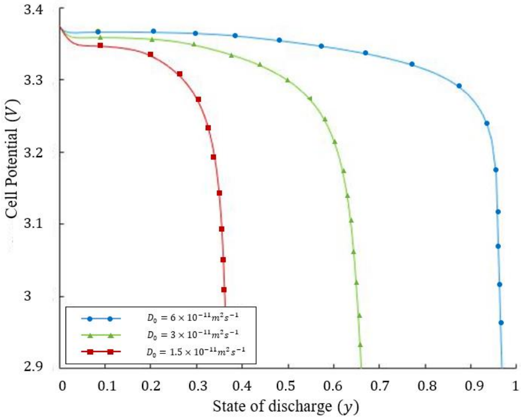

Figure 7. The discharge curve at 150 mAh/g discharge rate when diffusivity, D_0 is varied

5. CONCLUSION

This paper describes the charge transport mechanism of half-cell LiFePO$_4$ electrode material in LiPF$_6$ electrolyte. The effect of electrolyte parameter variation upon the performance of lithium ion batteries during discharge is then being analysed. The factors of limiting the discharge performance found to be the electrolyte depletion of Li^+ concentration and the fully discharge of particles that disallow the reaction occurs at electrode surface. The results show that the variation of electrolyte parameter influences the performance of the cells. The higher the diffusivity provides larger cell capacity as Li^+ is able to move back and forth easily in electrolyte and electrode particles. In addition, movement speed of lithium ion is depending on the concentration of the electrolyte. Higher initial concentration of lithium ion provides higher rate of reaction, hence, produce better cell discharge performance because it delays the electrolyte depletion to occur. Thus, varying the initial concentration and the diffusivity of Li^+ in electrolyte give significant influences to the performance of lithium ion battery.

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