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Theoretical Energy Density of Li-Air Batteries

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A model for predication of the gravimetric and volumetric energy densities of Li-air batteries using aqueous electrolytes is developed. The theoretical gravimetric/volumetric capacities and energy densities are calculated based on the minimum weight of the electrolyte and volume of air electrode needed for completion of the electrochemical reaction with Li metal as an anode electrode. It was determined that both theoretical gravimetric/volumetric capacities and energy densities are dependent on the porosity of the air electrode. For instance, at a porosity of 70%, the maximum theoretical cell capacities are 435 mAh/g and 509 mAh/cm³ in basic electrolyte, and 378 mAh/g and 452 mAh/cm³ in acidic electrolyte. The maximum theoretical cell energy densities are 1300 Wh/kg and 1520 Wh/L in basic electrolyte, and 1400 Wh/kg and 1680 Wh/L in acidic electrolyte. The significant deduction of cell capacity from specific capacity of Li metal is due to the bulky weight requirement from the electrolyte and air electrode materials. In contrast, the Li-air battery using a nonaqueous electrolyte does not consume electrolyte during the discharge process and has high cell energy density. For Li-air batteries using both aqueous and nonaqueous electrolytes, the weight increases by 8–13% and the volume decreases by 8–20% after the cell is fully discharged.

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The rapid growth of portable electronic devices in both military and civilian applications has driven a need for high-energy-density storage devices. Recently, lithium (Li)-air batteries have been attracting much attention due to its extremely high specific capacity. The reason for such high specific capacity is that there is a Li sheet as anode electrode and cathodic reactant of O₂ from air. The theoretical specific capacity of Li is 3862 mAh/g,¹ which is at least one order of magnitude higher than that of any type of electrode materials used in advanced Li-ion or Li-polymer batteries. Considering the high operational voltages 2.9–3.1 V for systems using nonaqueous electrolyte, the theoretical maximum energy density based on Li electrode can be over 11,000 Wh/kg, which is not only much higher than that of any advanced batteries but is also higher than that of fuel cells.

The concept of the Li-air battery was introduced by researchers at Lockheed who proposed the use of an aqueous alkaline solution as the electrolyte.² The Li-air batteries reduce oxygen from the environment at the air electrode surface to form an oxide. The operating voltage is 2.21 V. Abraham and Jiang reported a Li-air battery using nonaqueous electrolyte.³ The operating voltage of the batteries is 2.9–3.1 V and is higher than that of batteries using a aqueous electrolyte. However, due to the low oxygen solubility in nonaqueous electrolyte, the power density of Li-air battery using nonaqueous electrolytes is low.^{4,5} Recently, a Li-air battery that combines advantages from both batteries using aqueous and nonaqueous electrolytes was proposed.⁶ In the Li-air batteries, the nonaqueous electrolyte at the anode side is used in order to eliminate the dangerous reaction between metallic Li and water.

In most published papers, the theoretical maximum energy density of Li-air batteries was estimated based on Li anode electrode only and is extremely large; however, the practical discharge capacity and energy density of Li-air batteries are always cathode limited due to discharge products deposition and electrolyte consumption in the air electrode. In this paper, a model was developed for calculation of the gravimetric and volumetric energy densities. With this model, the maximum energy density and the optimized mass or volume ratio of Li anode and carbon air electrodes as well as electrolyte can be obtained. The influence of porosity of air electrode on

the energy density of the cell is discussed. The weight and volume changes of Li-air batteries during the discharge can also be obtained.

Model

The model for estimation of the gravimetric/volumetric capacities and energy densities of Li-air batteries is based on the cell structure shown in Fig. 1. It is a system based on the combination of aqueous and nonaqueous electrolytes. The anode electrode is a Li foil that provides the Li ion source. The nonaqueous electrolyte is used as a Li-ion-conducting medium for improving the interface between Li foil and membrane. The cathode (air) electrode is made with porous carbon and aqueous electrolyte, which filled the electrode pore volume. The role of carbon in the cathode electrode is to provide active sites for oxygen reduction reaction. The aqueous

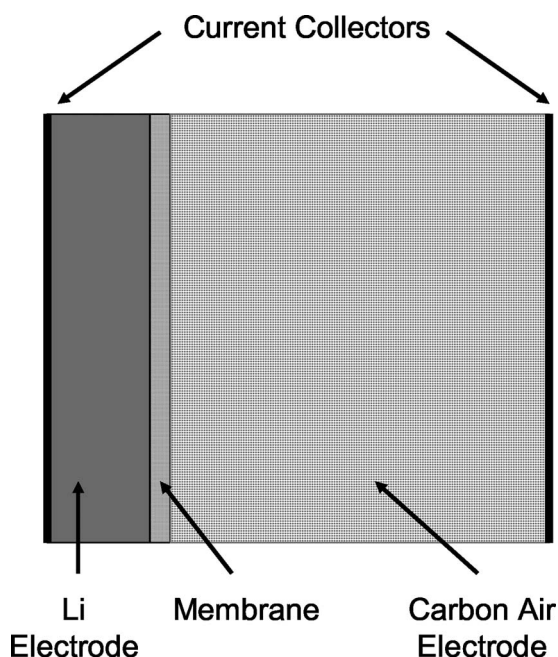


Figure 1. Schematic of a Li-air battery using a nonaqueous electrolyte at the anode side and aqueous electrolyte at the cathode side.

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electrolyte not only is a conducting medium for the Li ion but also provides the active anion during the oxidation reduction reaction. Therefore, the aqueous electrolyte in the cathode electrode is consumed during the discharge process and is considered as an active material in the cell along with Li and carbon at anode and cathode electrodes, respectively.⁷⁻¹⁰

The models for estimating the cell specific capacity and energy of Li-air batteries using aqueous electrolytes are developed. The theoretical maximum energy density and specific energy, the optimal mass/volume ratio, and the weight and volume changes after being fully discharged are calculated based on the following assumptions:

1. The gravimetric/volumetric capacities and energy densities are calculated based on weight or volume of the anode (Li) electrode, air electrode (carbon), and electrolyte only. Other materials, such as membrane, current collector, and package materials, are not included. Because the nonaqueous electrolyte at the anode electrode is not consumed during the charge and discharge processes, it is not included.

2. All pore volume in the air electrode (carbon) is accessible by electrolyte and solid Li precipitate from the reaction. After the cell is fully discharged, Li metal is 100% used and the pore volume in the air electrode is designed for just being able to be filled by solid Li precipitate.

3. After being fully discharged, no excess electrolyte is left in the cell.

The theoretical capacity and energy density of Li-air batteries is dependent on the material formation in the battery, particularly, the electrolyte used in the air-electrode, because the products due to the oxidation reaction during the discharge process are different and are determined by the electrolyte. Therefore, the calculation for capacity and energy density must be separated according to the electrolyte used in the air electrode.

Basic Electrolyte

When a basic electrolyte, such as diluted LiOH solution, is used in the air electrode, the overall reaction for Li-air cell at the air electrode side will be¹¹



From Eq. 1, it can be seen that 1 mole of Li will consume 1.5 mole of water from the electrolyte and 0.25 mole of oxygen from the air, then produce 1 mole of monohydrate lithium hydroxide ($\text{LiOH}\cdot\text{H}_2\text{O}$). From Eq. 1, it can be seen that the water is consumed during the cell discharge; therefore, the concentration of LiOH in the electrolyte increases.

The maximum specific capacity and energy will be achieved at a balance weight ratio according to Eq. 1, and the weight and volume of active materials including anode, cathode, and electrolyte are as follows:

1. Anode electrode (Li metal): The weight of Li metal is m_L . The specific capacity of anode electrode is

$$c_L = \frac{F}{w_L} \quad [2]$$

where $F = 96,484 \text{ C/mol}$ is the Faraday constant and $w_L = 6.941 \text{ g/mol}$ is the atomic weight of Li. The specific capacity of the Li electrode is 3861 mAh/g . The volume of the anode electrode is

$$v_L = \frac{m_L}{\rho_L} \quad [3]$$

where $\rho_L = 0.535 \text{ g/cm}^3$ is the mass density of Li.

2. Electrolyte: The weight of the electrolyte (water) is

$$m_E = \frac{3}{2} m_L w_{\text{HO}} \quad [4]$$

where $w_{\text{HO}} = 18 \text{ g/mol}$ is the molecular weight of water. The volume of electrolyte is

$$v_{\text{HO}} = \frac{m_E}{\rho_{\text{HO}}} \quad [5]$$

where $\rho_{\text{HO}} = 1 \text{ g/cm}^3$ is the mass density of water.

3. Li precipitate of $\text{LiOH}\cdot\text{H}_2\text{O}$: The weight of $\text{LiOH}\cdot\text{H}_2\text{O}$ is

$$m_{\text{LOH}} = \frac{m_L}{w_L} w_{\text{LOH}} \quad [6]$$

where $w_{\text{LOH}} = 41.96 \text{ g/mol}$ is the molecular weight of $\text{LiOH}\cdot\text{H}_2\text{O}$. The volume of $\text{LiOH}\cdot\text{H}_2\text{O}$ is

$$v_{\text{LOH}} = \frac{m_{\text{LOH}}}{\rho_{\text{LOH}}} \quad [7]$$

where $\rho_{\text{LOH}} = 1.51 \text{ g/cm}^3$ is the mass density of $\text{LiOH}\cdot\text{H}_2\text{O}$.

4. Air electrode: The volume of air electrode is

$$v_C = \frac{v_{\text{LOH}}}{p} \quad [8]$$

where p is the porosity of the air electrode. The weight of carbon is

$$m_C = v_C \rho_C (1 - p) \quad [9]$$

where $\rho_C = 2.26 \text{ g/cm}^3$ is the mass density of bulk carbon.

The cell specific capacity of Li-air batteries using basic electrolyte is

$$c_p = \frac{C_L}{m_L + m_{\text{HO}} + m_C} = \frac{F}{w_L + (3/2)w_{\text{HO}} + (w_{\text{LOH}}/\rho_{\text{LOH}})\rho_C[(1-p)/p]} \quad [10]$$

where $C_L = c_L m_L$ is the capacity of the Li electrode.

It should be noted that the specific capacity described above is based on the initial weight of the active materials; however, the weight of the cell changes after discharge. From Eq. 10, it can be seen that the theoretical cell specific capacity is only a function of porosity of the air electrode. From Eq. 1, the molecule ratio of H_2O to $\text{LiOH}\cdot\text{H}_2\text{O}$ is 1.5. It is found that the volume for 1 mole of $\text{LiOH}\cdot\text{H}_2\text{O}$ is >1.5 mole of water by a factor of $\sim 3\%$. Excess electrolyte (water) is needed in order to fill the pore volume in the air electrode; therefore, a correction factor will be applied to Eq. 10 as

$$c_p = \frac{F}{w_L + (w_{\text{LOH}}/\rho_{\text{LOH}})\rho_{\text{HO}} + (w_{\text{LOH}}/\rho_{\text{LOH}})\rho_C[(1-p)p]} \quad [11]$$

The first term in the denominator is the weight of 1 mole of Li in an anode electrode. The second and third terms in the denominator are the necessary weight of water and carbon material electrode in an air electrode for 1 mole of Li used in an anode electrode, respectively. The volumetric capacity can be expressed as

$$c_v = \frac{C_L}{v_L + v_C} = \frac{F}{(w_L/\rho_L) + (w_{\text{LOH}}/\rho_{\text{LOH}})(1/p)} \quad [12]$$

Figure 2 shows the theoretical specific capacity and volumetric capacity as a function of porosity of the air electrode. It can be seen that the capacity increases with increasing the porosity of the air electrode. At a porosity of 70%, the specific capacity and volumetric capacity are 435 mAh/g and 509 mAh/cm^3 , respectively.

The energy density of Li-air batteries using a basic electrolyte is determined by the specific capacitance and the cell voltage. It is assumed that the voltage of the cell is a constant during the entire discharge process and is determined by the following reactions:^{1,12}

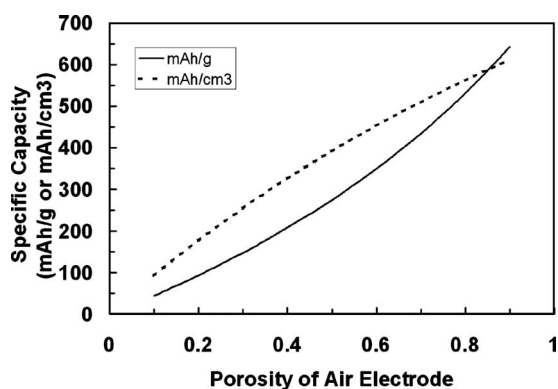


Figure 2. Theoretical specific and volumetric capacities of a Li-air battery using a basic electrolyte based on Li electrode, air electrode, and electrolyte as a function of porosity of the air electrode.

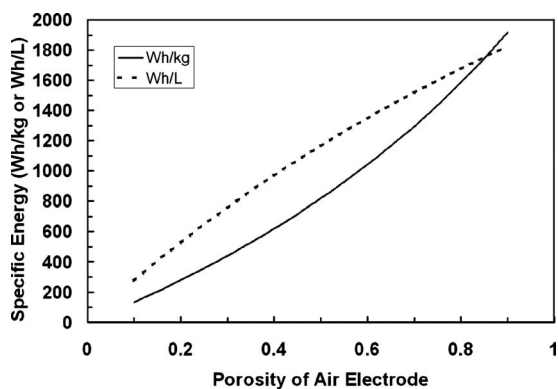
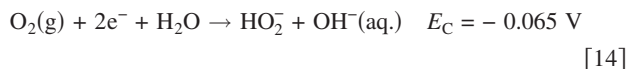


Figure 3. Specific energy and energy density as a function of porosity of the air electrode before discharge. The electrolyte is a basic solution.

At anode



At cathode (two-electron reaction)



The cell voltage

$$V_o = E_C - E_A = 2.982 \text{ V} \quad [15]$$

The specific energy

$$\varepsilon_p = c_p V_o \quad [16]$$

The energy density

$$\varepsilon_v = c_v V_o \quad [17]$$

Figure 3 shows the theoretical specific energy and energy density as a function of porosity of the air electrode. At the porosity of 70%, the specific energy and energy density are 1300 Wh/kg and 1520 Wh/L, respectively.

In order to achieve the maximum specific capacity and energy, the mass ratio will be optimized according to the mole ratio as expressed in Eq. 1. Figure 4 shows the mass of the electrolyte and carbon electrode per gram of Li metal. Correspondingly, the volume of electrolyte and electrode materials per gram of Li metal is shown in Fig. 5.

Both weight and volume changes during the discharge process are also calculated. Figure 6 shows the weight and the volume ratios between a fresh cell and fully discharged cell. It can be seen that at a porosity of 70%, the weight increases 13% and the volume de-

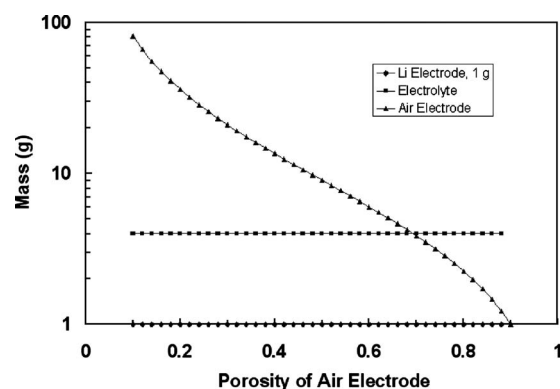


Figure 4. The optimal mass for the electrolyte and air electrode based on 1 g of Li metal as a function of porosity of the air electrode. The electrolyte is a basic solution.

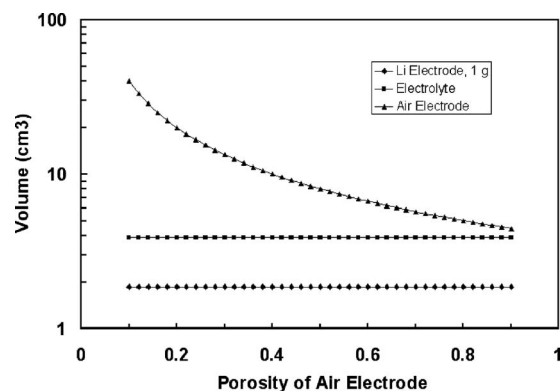


Figure 5. The volume of Li and air electrodes, and electrolyte based on 1 g of Li metal as a function of porosity of the air electrode. The electrolyte is a basic solution.

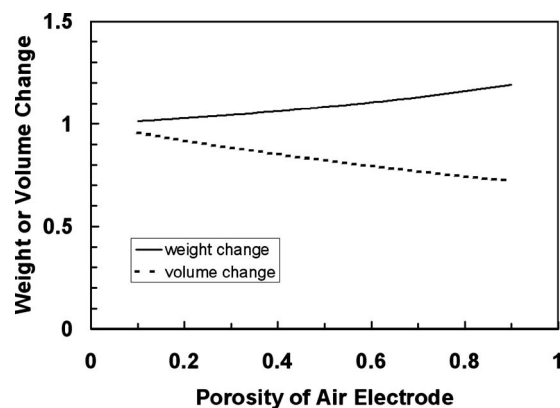
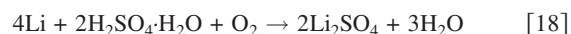


Figure 6. The weight and volume ratios between a fresh cell and fully discharged cell as a function of porosity of the air electrode. The electrolyte is a basic solution.

creases 23% after discharge. Therefore, a mechanical mechanism, such as a spring, may be needed for applying a pressure to the vertical direction of electrode plates.

Acidic Electrolyte: $\text{H}_2\text{SO}_4 \cdot \text{H}_2\text{O}$

Overall the reaction for Li-air cell using acidic electrolyte will be



It can be seen that 1 mole of Li will consume 0.5 mole of monohydrate sulfuric acid molecules from the electrolyte and 0.25 mole of oxygen from the air, and then produce 1 mole of anhydrous lithium sulfate (Li_2SO_4) as a precipitate. The calculation is based on Eq. 18 and assumes that dehydrated lithium sulfate is formed as the reaction precipitate; however, the specific capacity and energy will not change if the hydrate lithium sulfate is formed as well as the amount of water is < 1.5 mole in each mole of lithium sulfate structure.

For Li-air batteries using acidic electrolyte, in addition to Eq. 2 and 3 for calculation of the capacity and volume of anode electrode, the following equations for weight and volume are used during the calculation:

1. Electrolyte: The weight of electrolyte is

$$m_{\text{HSO}} = \frac{1}{2} \frac{m_{\text{L}}}{w_{\text{L}}} w_{\text{HSO}} \quad [19]$$

where $w_{\text{HSO}} = 116.09$ g/mol is the molecular weight of $\text{H}_2\text{SO}_4 \cdot \text{H}_2\text{O}$. The volume of electrolyte is

$$v_{\text{HSO}} = \frac{m_{\text{HSO}}}{\rho_{\text{HSO}}} \quad [20]$$

where $\rho_{\text{HSO}} = 1.788$ g/cm³ is the mass density of $\text{H}_2\text{SO}_4 \cdot \text{H}_2\text{O}$.¹

2. Li precipitate of Li_2SO_4 : The weight of Li_2SO_4 is

$$m_{\text{LSO}} = \frac{1}{2} \frac{m_{\text{L}}}{w_{\text{L}}} w_{\text{LSO}} \quad [21]$$

where $w_{\text{LSO}} = 109.94$ g/mol is the molecular weight of Li_2SO_4 . The volume of Li_2SO_4 is

$$v_{\text{LSO}} = \frac{m_{\text{LSO}}}{\rho_{\text{LSO}}} \quad [22]$$

where $\rho_{\text{LSO}} = 2.221$ g/cm³ is the mass density of Li_2SO_4 .

3. Air electrode: Because the volumes of Li_2SO_4 , $\text{H}_2\text{SO}_4 \cdot \text{H}_2\text{O}$, and H_2O have the following relationship as

$$v_{\text{LSO}} < v_{\text{HSO}} < v_{\text{HSO}} + v_{\text{H}_2\text{O}}$$

where $v_{\text{H}_2\text{O}}$ is the volume of water. The volume of the air electrode is determined by the volume of $\text{H}_2\text{SO}_4 \cdot \text{H}_2\text{O}$ electrolyte as

$$v_{\text{C}} = \frac{v_{\text{HSO}}}{p} \quad [23]$$

where p is porosity of the air electrode. The weight of carbon is

$$m_{\text{C}} = v_{\text{C}} \rho_{\text{C}} (1 - p) \quad [24]$$

where $\rho_{\text{C}} = 2.26$ g/cm³ is the mass density of carbon.

The cell specific capacity can be expressed as

$$c_{\text{p}} = \frac{C_{\text{L}}}{m_{\text{L}} + m_{\text{HSO}} + m_{\text{C}}} = \frac{F}{w_{\text{L}} + (1/2)w_{\text{HSO}} + (1/2)(w_{\text{HSO}}/\rho_{\text{HSO}})\rho_{\text{C}}[(1-p)/p]} \quad [25]$$

The volumetric capacity is

$$c_{\text{v}} = \frac{C_{\text{L}}}{v_{\text{L}} + v_{\text{C}}} = \frac{F}{(w_{\text{L}}/\rho_{\text{L}}) + (1/2)(w_{\text{HSO}}/\rho_{\text{HSO}})(1/p)} \quad [26]$$

Figure 7 shows the theoretical specific capacity and volumetric capacity as a function of porosity of the air electrode. It can be seen that capacity increases with increasing the porosity of the air electrode and at a porosity of 70%, the specific capacity and volumetric capacity are 378 mAh/g and 452 mAh/cm³, respectively.

The energy density for using the acidic electrolyte can also be calculated based on the specific capacity and cell voltage.

At anode

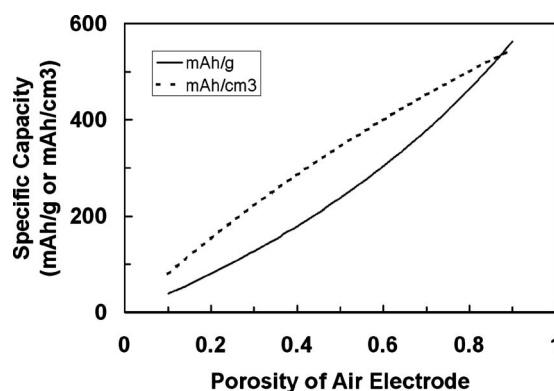


Figure 7. Theoretical specific and volumetric capacities of the Li-air battery using H_2SO_4 solution as the electrolyte calculated based on anode and air electrodes, and electrolyte as a function of porosity of the air electrode.



At cathode (two electron reaction)



The cell voltage

$$V_{\text{o}} = E_{\text{C}} - E_{\text{A}} = 3.72 \text{ V} \quad [29]$$

The specific energy

$$\varepsilon_{\text{p}} = c_{\text{p}} V_{\text{o}} \quad [30]$$

The energy density

$$\varepsilon_{\text{v}} = c_{\text{v}} V_{\text{o}} \quad [31]$$

Figure 8 shows the theoretical specific energy and energy density as a function of porosity of the air electrode. At a porosity of 70%, the specific energy and energy density are 1400 Wh/kg and 1680 Wh/L, respectively.

Figure 9 shows the mass of the electrolyte and air electrode per gram of Li metal. Correspondingly, the volume of electrolyte and electrode materials per gram of Li metal is shown in Fig. 10.

Figure 11 shows weight and volume ratios between a fresh cell and a fully discharged cell. It can be seen that at a porosity of 70%, the weight increases 8.3% and the volume decreases 20% after discharge.

Organic Electrolyte

As a comparison, the theoretical gravimetric and volumetric capacities and energy densities for Li-air cell using nonaqueous electrolyte is also calculated. The overall reaction for Li-air cell using nonaqueous electrolyte solution can be expressed as

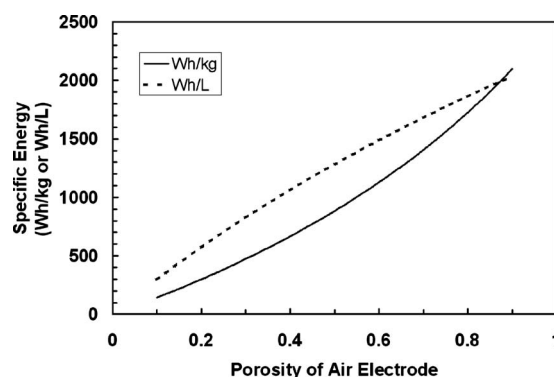


Figure 8. Specific energy and energy density as a function of porosity of the air electrode before discharge. The electrolyte is a H_2SO_4 solution.

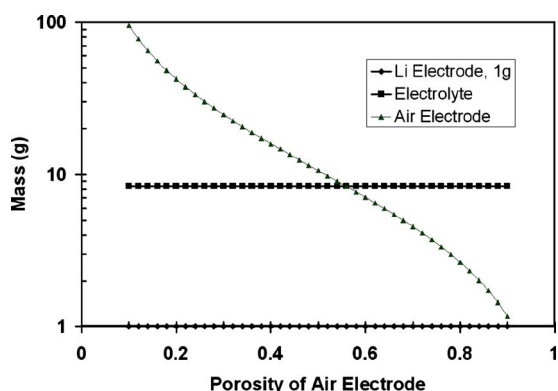


Figure 9. (Color online) The optimal mass for the electrolyte and air electrode based on 1 g of Li metal as a function of porosity of the air electrode. The electrolyte is a H_2SO_4 solution.

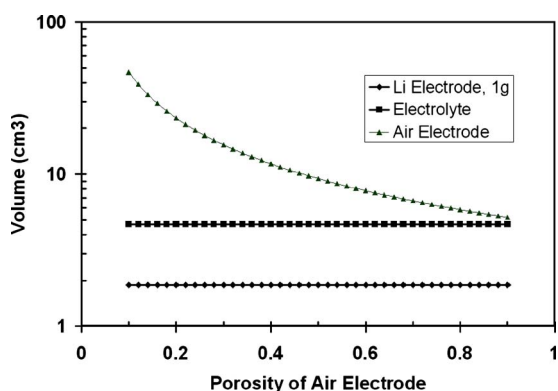


Figure 10. (Color online) The volume of Li and air electrodes, and electrolyte based on 1 g of Li metal as a function of porosity of the air electrode. The electrolyte is a H_2SO_4 solution.

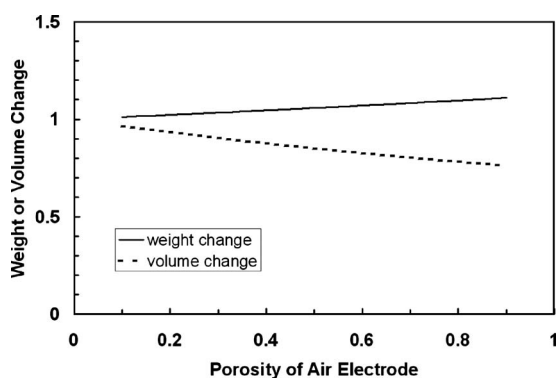


Figure 11. The weight and volume ratios between a fresh cell and fully discharged cell as a function of porosity of the air electrode. The electrolyte is a H_2SO_4 solution.



It can be seen that 1 mole of Li will consume 0.5 mole of oxygen from the air, then produce 0.5 mole of lithium peroxide (Li_2O_2) as a precipitate at the air electrode. No electrolyte is consumed during the discharge process; however, a minimum amount of electrolyte is needed to fill up the pore volume in the air electrode and provide a Li ion conductive path from the anode electrode to the air electrode. The electrolyte is 1 M LiPF_6 in a propylene carbonate/dimethyl carbonate (50/50) solution. The following parameters are used during

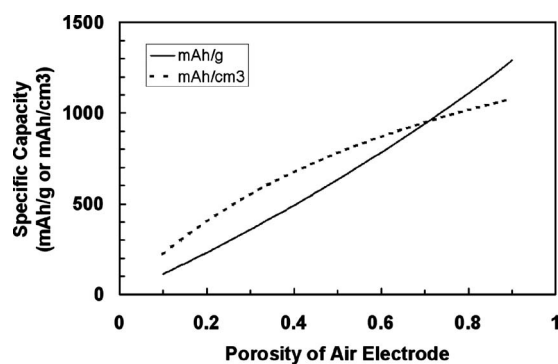


Figure 12. Theoretical specific and volumetric capacities of the Li-air battery using a nonaqueous electrolyte as a function of porosity of the air electrode.

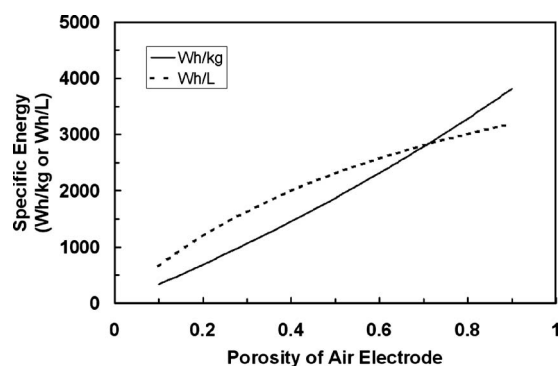


Figure 13. Specific energy and energy density as a function of porosity of the air electrode before discharge. The electrolyte is a nonaqueous electrolyte.

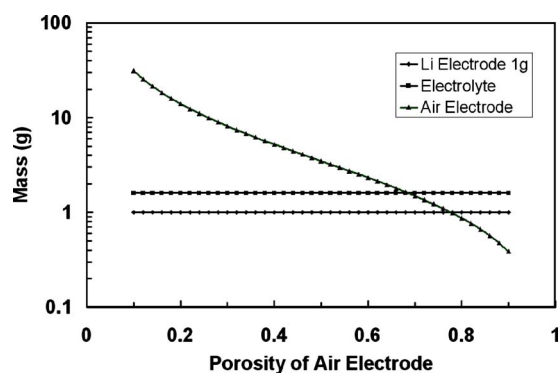


Figure 14. (Color online) The optimal mass for electrolyte and the air electrode based on 1 g of Li metal as a function of porosity of the air electrode. The electrolyte is a nonaqueous electrolyte.

the calculation: The mass density and molecular weight of Li_2O_2 are 2.14 g/cm^3 and 45.88 g/mol ; the mass density of electrolyte is 1.0402 g/cm^3 ; and the cell voltage is 2.959 V . This follows the same calculation procedure as used for aqueous electrolytes; the gravimetric and volumetric capacities, energy densities, mass of two electrodes and electrolyte, volume of two electrode and electrolyte, and volume change after the discharge can be obtained and is shown in Fig. 12-16, respectively. It can be seen that at a porosity of 70% for the air electrode, the cell capacities are 940 mAh/g and 950 mAh/cm^3 , and energy densities are about 2790 Wh/kg and

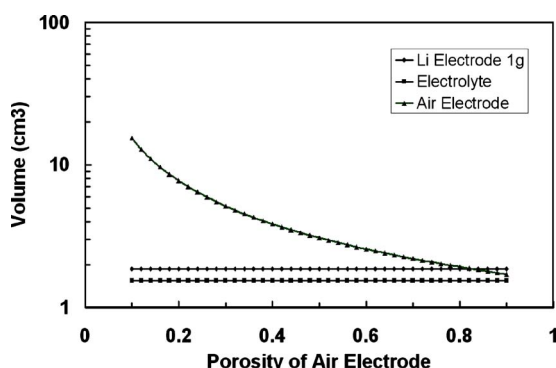


Figure 15. (Color online) The volume of the anode and the air electrodes, and electrolyte based on 1 g of Li metal as a function of porosity of the air electrode. The electrolyte is a nonaqueous electrolyte.

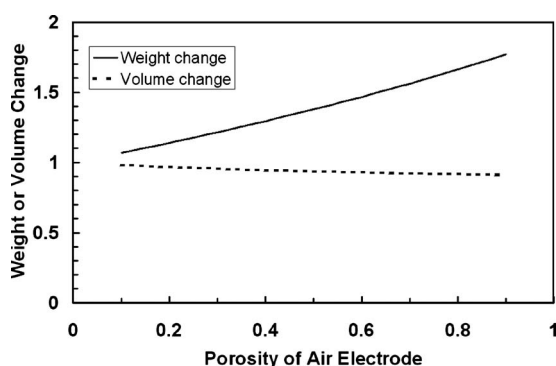


Figure 16. The weight and volume ratios between a fresh cell and fully discharged cell as a function of porosity of the air electrode. The electrolyte is a nonaqueous electrolyte.

2800 Wh/L, which is higher than that using aqueous electrolytes.

Discussion

The above specific capacity and energy projections are the theoretical maximum limitation and are based on active materials including only Li metal, air electrode, and electrolyte. It is assumed that the cell is assembled in a way that the weight and volume for active materials are perfectly balanced according to the electrochemical reaction. However, in practical cells, some excess electrolyte must be in the cell in order to provide ionic conductive media during the entire discharge process. In addition, other necessary materials, including current collectors, membrane, and package materials, will further reduce the cell specific capacity and energy by 20–30%.

One big assumption made in this model is that the pore volume in the air electrode is completely filled by the reaction products after

being fully discharged. But, in reality, some pore volume, particularly, some pores, may not be filled because they may have a smaller volume than the ions needed to fill these pores. How much deduction due to this fact can only be determined by experiments.

The cell specific capacity and energy for the cell using aqueous electrolytes are about half of that using nonaqueous electrolytes. This is because for the cells using aqueous electrolytes, the electrolyte is consumed and the precipitate is bulky and heavy. The weight of the electrolyte needed in the cell using aqueous electrolytes is two to five times heavier than that using nonaqueous electrolytes. The weight and volume of the air electrodes are also about four times greater for using aqueous electrolytes than nonaqueous electrolytes.

The cell specific capacity and energy are strongly dependent on the porosity of the air electrode and increase with increasing the porosity. The porosity of the air electrode is dependent on the type of carbon used. Usually, high porosity or low density of carbon electrode may result in low electrical conductance, which will affect the power performance of the cell. Therefore, air electrodes with high porosity and high electrical conductivity is crucial for achieving high energy density of Li-air batteries.

Conclusion

The maximum possible gravimetric and volumetric energy densities of Li-air batteries using aqueous and nonaqueous electrolytes was calculated based on charge balance theory. It is found that the maximum energy density of Li-air batteries is strongly dependent on the porosity of the air electrode and the electrolyte used at the cathode side. The theoretical energy density of Li-air batteries using aqueous electrolytes is considerably less than that using a nonaqueous electrolyte. The model can be used not only for predicting the maximum energy density but also for designing a battery with optimal mass ratios between two electrodes and electrode to electrolyte.

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