Optimization of CNT/Activated Carbon-Based Cathodes for High-Performance Aluminum-Air Batteries

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Abstract: We developed a high-performance air-cathode for aluminum-air batteries based on carbon nanotube/activated carbon material loading in a carbon sheet substrate. By using activated carbon, which has a large surface area, the air-cathode performance can be improved. Carbon nanotube coating liquid plays the role of a binder to bind activated carbon powder to the substrate. The fabricated air-cathode was optimized to obtain the maximum power density. The maximum power density was 2.3mW/cm^2 obtained at 7.9mA/cm^2 current density, respectively.

Key words: Aluminum-air battery, activated carbon, carbon nanotube, solid electrolyte.

1. Introduction

Researchers have been studying the development of new energy sources. The main reason is the exhaustion of resources and the environmental impact. Rechargeable batteries have received the attention of many researchers. Among them, lithium batteries have been extensively studied. However, lithium is an expensive metal. Material cost is an important factor which limits the spreading of lithium batteries.

In this research, we focused on using aluminum as a material with high energy density instead of lithium. Aluminum is the most abundant metal in the crust. This advantage along with its low price-per–energy unit have increased interest in its usage as an anode material in a metal-air battery system, specifically, in alkaline metal-air batteries [1]. The aluminum–air battery has many advantages, such as abundance, low cost, innocuousness, high power density, high energy density, etc [2]. The aluminum-air battery possesses high theoretical specific energy capacity (2980 Ahkg^{-1}), which is the second highest after that of lithium (3860 Ahkg^{-1}) and is capable of outputting high current densities [3].

The cathode has a great influence on the performance of aluminum-air batteries. Research on carbon nanotube (CNT) has been focused on its properties of high conductivity and high specific surface area. There are two types of CNTs: single-walled CNT (SWCNT) and multi-walled CNT (MWCNT) [4]-[6]. SWCNT consists of a single graphene layer with high conductivity. However, it is expensive and therefore, difficult for mass adoption. MWCNT consists of multiple graphene layers. Although it has been reported that the performance of MWCNT is inferior to SWCNT, MWCNT is advantageous in terms of cost and availability.

Because high specific surface area and simple preparation process, activated carbon has been extensively used in adsorption application, hydrogen storage, and supercapacitors [7], [8]. Activated carbon is made from natural materials, so using activated carbon for electrical components is environment-friendly. Activated carbon-based air–cathode is such a robust and highly efficient air-cathode with advantages of low cost (30\$$m^{-2}$ compared to 1600\$$m^{-2}$ of Pt–Nafion cathode) and high reproducibility [9]-[11].

In this paper, to increase the specific surface area of the air-cathode of the aluminum-air battery, we utilized MWCNT mixed with activated carbon loading on a carbon sheet substrate to make the air-cathode. One purpose of this research was to create a high-performance CNT/activated carbon-based cathode by a simple fabrication method. Also, the electrode was optimized to obtain the highest power generation.

2. Experiment

2.1. Measurement Setup

The experimental setup is shown in Fig. 1. We used a digital multimeter (PEAK METER, MS 8233D) for this measurement of the output voltage. We measured the discharging voltage over several load resistors. The load resistances used to measure the power density were 10kΩ, 5kΩ, 1kΩ, 460Ω, 323Ω, 100Ω, 50Ω, 10Ω, 1Ω. Current density and power density were calculated based on the measured discharging voltage.

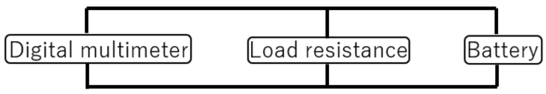


Fig. 1. Experimental setup.

2.2. Battery Design

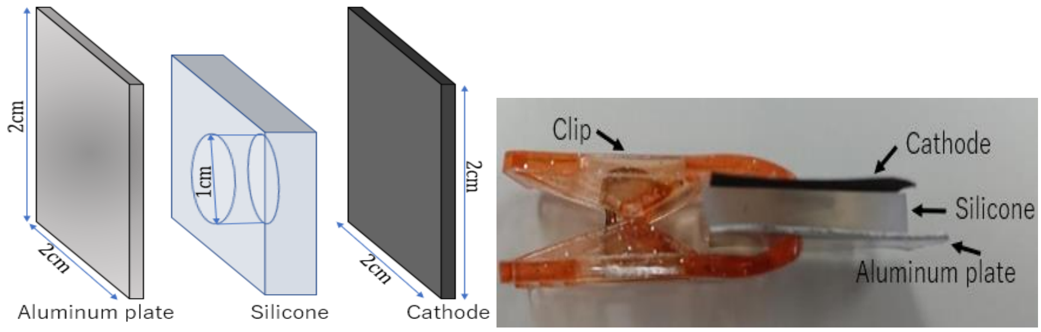


Fig. 2. Battery design.

The design of the battery is shown in Fig. 2. An aluminum plate (Hikari Yunihobi, HA 2412) was used as the anode. The film thickness was 0.2mm, and the size was 2×2cm. We prepared an aqueous solution of sodium chloride having a mass concentration of 10% and then the solution was gelled by adding a water absorbent polymer (Newstone International Corporation Tokyo, Japan) to make a gel electrolyte. A silicone film (film thickness, 3mm) was cut into 2×2cm and drilled a hole of 10mm in diameter. The hole was the place where holding gel electrolyte. The cathode size was also 2×2cm. The electrolyte gel was put in the hole of the silicone case. A clip was used to hold the anode and cathode to the battery case.

2.3. Cathode Fabrication

We prepared a carbon-based cathode made by loading CNT/activated carbon on a carbon sheet(AZUMI FILTERPAPER CO., Ltd). The carbon sheet consisted of 50% carbon fiber, 50% cellulose, and the thickness was 0.2mm, the volume resistivity was 109mΩ・cm. Activated carbon powder was made from coconut husk (USE Co., Ltd. KD-PWSP-1, particle size, 6μ).

Activated carbon powder and water were mixed in a beaker by a magnetic stirrer (AS ONE, REXIM RS-6 DN) for 5h at the speed of 1500rpm. CNT coating liquid (N7006L, KJ specialty paper, Japan) was dispersed by an ultrasonic homogenizer for 30min (TOMY, UD-211, LST-100) and then CNT was added to the beaker. The mixture was mixed for 24h at the speed of 1500rpm. Using this method, we prepared five patterns of CNT/activated carbon mixtures with different amount of activated carbon powder as listed in Table 1.

To make the cathode, carbon sheets were dipped in the prepared solutions for 1 minute. Then they were taken out for drying in an electric oven at 60°C for 30 minutes. Five samples of the cathodes were named AC1 - AC5 as in Table 1.

Table 1. Amount of Activated Carbon

Activated carbon (g)	Water (ml)	CNT (ml)	Sample
0.5	20	3	AC1
1.0	20	3	AC2
2.0	20	3	AC3
2.5	20	3	AC4
3.0	20	3	AC5

2.4. SEM Images

We used a scanning electron microscopy (SEM, S-4300 Hitachi) to observe the surface of the original carbon sheet and the cathodes with different amount of activated carbon powder (AC1 – AC5).

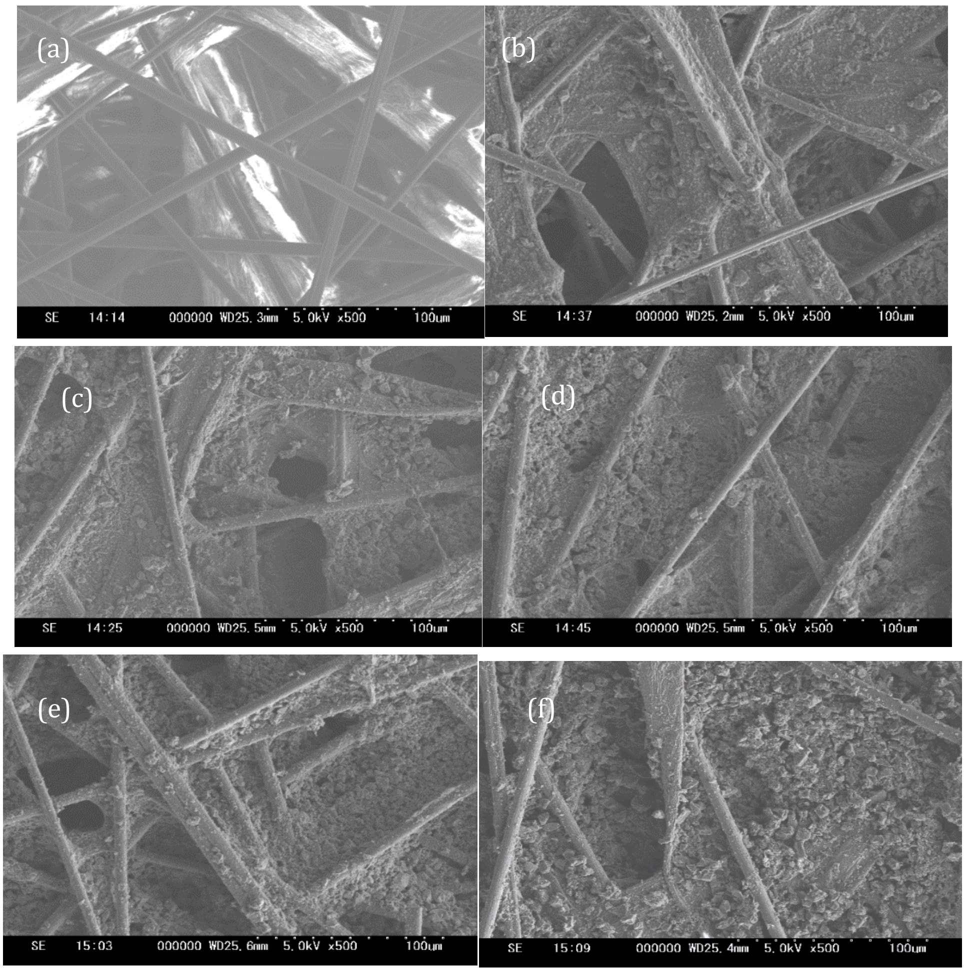


Fig. 3. SEM images. (a) Original carbon sheet, (b) AC1, (c) AC2, (d) AC3, (e) AC4, (f) AC5.

3. Result

3.1. SEM Images

The surfaces of the original carbon sheet (before loading CNT/activated carbon mixtures) and AC1 – AC5 cathode electrodes were observed by SEM. Fig. 3 shows the SEM images. Fig. 3(a) shows the image of the original carbon sheet. Thin fibers of carbon and cellulose were entangled with multiple layers and many empty spaces. Fig. 3(b) –Fig. 3(f) were the images of the cathode electrodes AC1 – AC5, respectively. A big difference was observed between the surfaces of AC1 and AC3. There was little CNT/activated carbon covered on the surface of AC1.

On the other hand, there was almost entirely CNT/activated carbon covered on the surface of AC3. Comparing the surfaces of AC3 – AC5, there was little difference between them. This result showed that, based on this fabrication method, we had reached the maximum loading capacity of CNT/activated carbon in these electrodes.

3.2. Battery Measurement

In this paper, we studied the difference in the characteristics of the batteries using different air-cathodes: AC1 - AC5.

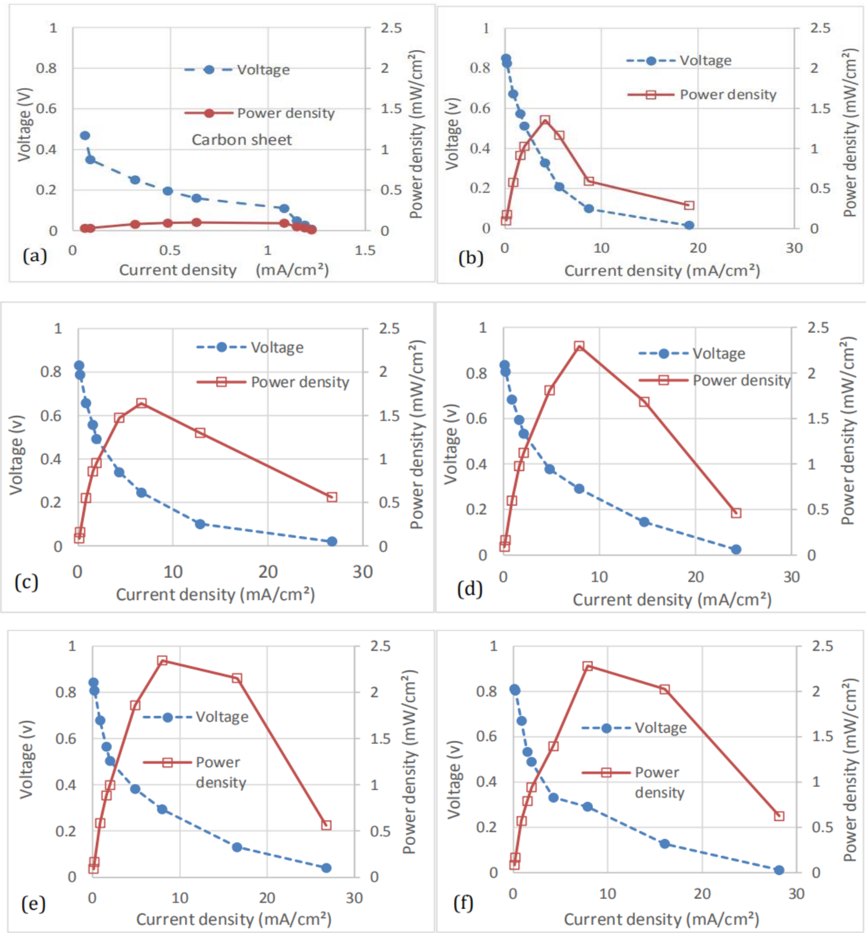
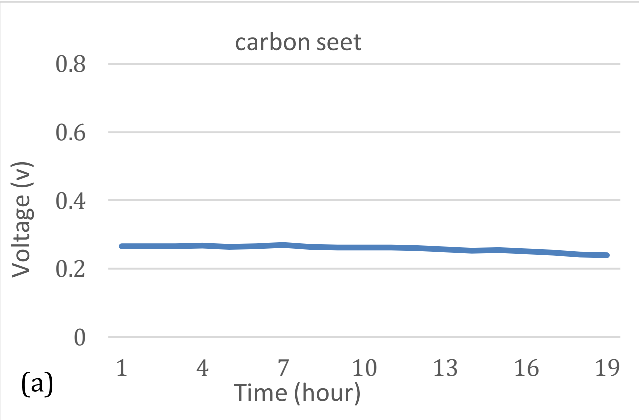


Fig. 4. The discharging voltage and power density of the batteries using (a) Carbon sheet, (b) AC1, (c) AC2, (d) AC3, (e) AC4, and (f) AC5.

Fig. 4 (a) - (f) show the discharging voltage and power density of batteries using carbon sheet, AC1 – AC5 air-cathodes, respectively. The discharging voltages were measured at 10kΩ, 5kΩ, 1kΩ, 460Ω, 323Ω, 100Ω, 50Ω, 10Ω, 1Ω external resistances; and the power densities were calculated based on these discharging voltages. It can be seen from the graphs (Fig. 4(b) –Fig. 4(d)) that the output power was significantly improved from 1.35 to 2.29mW/cm^2 (about 70%) as the concentration of activated carbon in table 1 increased from 0.5g to 2.0g (AC1 – AC3), respectively. This was because of the improvement of the specific surface area of the air-cathode. However, as the amount of activated carbon increased from 2.0g to 3.0g (AC3 – AC5), the results were almost the same with the power density around 2.3mW/cm^2 (Fig. 4(d) –Fig. 4(f)). This was attributed to the amount of CNT/activated carbon loaded in AC3 – AC5 was almost the same (Fig. 3(d) –Fig. 4(f)). The internal resistance of this battery was estimated around 50Ω. The maximum power density of the battery was about 2.3mW/cm^2 at about 7.9mA/cm^2 current density.



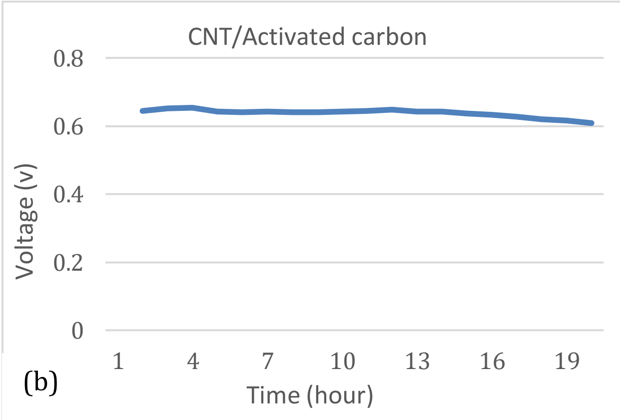


Fig. 5. Discharging voltage of the battery. (a) carbon sheet for the cathode. (b) CNT / activated carbon for cathode.

Fig. 5 shows the voltage transition of using carbon sheet or CNT / activated carbon for the cathode. We can confirm that CNT / activated carbon is better than the carbon sheet for the cathode.

4. Conclusion

In this research, we focused on developing a high-performance aluminum-air battery with the characteristics of easy to use, low cost, and safety. Therefore, CNT/activated carbon-loaded cathode electrodes were used to improve the performance of aluminum-air batteries. The experimental results showed that the more CNT/activated carbon loaded in the cathode, the higher the output power density. By optimizing the amount of activated carbon powder, the battery power density was increased by about 70% with a maximum of about 2.3mW/cm^2.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

Tatsuya, Kozo conducted the research; Kozo, Trang analyzed the data; Trang, Kozo wrote the paper; all authors had approved the final version.

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