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An Integrated Device of a Lithium-Ion Battery Combined with Silicon Solar Cells

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Abstract: This study reports an integrated device of a lithium-ion battery (LIB) connected with Si solar cells. A $\text{Li}(\text{Ni}_{0.65}\text{Co}_{0.15}\text{Mn}_{0.20})\text{O}_2$ (NCM) cathode and a graphite (G) anode were used to fabricate the lithium-ion battery (LIB). The surface and shape morphologies of NCM and graphite powder were characterized by field emission scanning electron microscopy (FE-SEM). The structural properties of NCM and graphite powder were determined by X-ray diffraction (XRD) analysis. XRD patterns of powders were well matched with those of JCPDS data. To investigate the electrochemical characteristics of NCM and graphite, cycling tests were performed after assembling the NCM-Li, the G-Li half-cell, and the NCM-G full-cell. The discharge capacity of the NCM cathode at 0.1C was 189.82 mAh/g⁻¹. The NCM-graphite full-cell showed 98.25% cycle retention at 1C after 50 cycles. To obtain enough charging voltage for the LIB connected with solar cells in an integrated device, eight single Si solar cells were connected in a series. The short-circuit photocurrent density for Si solar cells was 4.124 mA/cm². The fill factor and the open circuit voltage were 0.78 and 4.5 V, respectively. These Si solar cells showed a power conversion efficiency of 14.45%. The power conversion and storage efficiency of the integrated device of the NCM battery and Si solar cells was 7.74%. Charging of the integrated device could be as effective as charging with a battery cyclers.

Keywords: $\text{Li}(\text{Ni}_x\text{Co}_y\text{Mn}_z)\text{O}_2$; graphite; lithium-ion battery; silicon solar cell; integrated device



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1. Introduction

As the industry develops and the population grows, the world's energy demand has been increasing every year. Fossil fuels supply most of the global energy needs. In recent years, problems of depletion of fossil fuels and environmental pollution such as SO_x and NO_x emission problems caused by wastes of fossil fuels have emerged around the world [1]. To solve these problems, interest in renewable energy has increased. Among renewable energy sources, sunlight is the most abundant one. It is more widely used than other renewable energies such as geothermal, wind, and tidal power [2]. However, sunlight is highly dependent on weather conditions. In addition, it cannot be used at night. To use sunlight effectively, it is necessary to store solar energy in a battery or a grid [3,4].

Many research studies have reported that a converter is necessary to charge from solar energy to a battery system. However, the system that uses a converter could be more complex and expensive than a direct connection between a solar cell and a battery system [5–9]. In addition, it can be less efficient than direct charging a battery with solar energy [10]. Therefore, a direct DC current storing is in the spotlight as a promising technology using an integrated method [11].

Lithium-ion batteries (LIBs) have been applied to portable devices and electric vehicles (EV). Conventionally, LIBs are manufactured using lithium metal oxide cathode electrode materials such as LiCoO_2 (LCO), LiFePO_4 (LFP), $\text{Li}(\text{Ni}_x\text{Co}_y\text{Al}_z)\text{O}_2$ (NCA), and $\text{Li}(\text{Ni}_x\text{Co}_y\text{Mn}_z)\text{O}_2$ (NCM) with anode electrode materials [12,13].

Among various cathode materials, LFP has the advantages of a low price and high safety. Therefore, LFP is generally widely used in energy storage systems (ESSs). However, LFP has the disadvantages of low electronic conductivity and capacity [14,15]. NCA has a high energy density and high power. Thus, it has a higher capacity than LFP. However, NCA has a disadvantage in that its capacity retention rate is low, and its performance is degraded when stored at high temperatures [16]. Although NCM has similar properties to NCA, NCM has a higher cycle performance and C rates than those of NCA [17].

In this study, an integrated device of LIB connected Si solar cells was developed. The shape and structures of the powder of the NCM622 cathode and the graphite anode material were evaluated by field-emission scanning electron microscopy (FE-SEM) and X-ray diffraction (XRD) analysis, respectively. Electrochemical characteristics were evaluated after assembling NCM, the graphite half-cell, and the NCM-G full-cell. To obtain enough charging voltage for the battery, eight single Si solar cells in a series were connected. Electrical properties such as the fill factor (FF), the short circuit photocurrent density (I_{SC}), and the open circuit voltage (V_{OC}) were analyzed with a solar simulator. To develop an integrated device, Si solar cells and NCM-graphite LIB were combined. Photo-charging from solar cells to the NCM battery and galvanostatic charging from a cyclizer to the NCM battery were compared.

2. Materials and Methods

2.1. Preparation of LIB and Solar Cell

A cathode electrode slurry was prepared by adding NCM powder, polyvinylidene difluoride (PVDF), and carbon black (super P) to N-Methyl-2-pyrrolidone (NMP) solvent at a weight percent ratio of 8:1:1. The anode electrode slurry was made using graphite powder in the same way. Cathode and anode slurries were coated on Al foil and Cu foil, respectively. The coated slurry was dried and pressed to make a sheet electrode. The half-cell and the full-cell were assembled in a glove box. As an electrolyte, 1.2 M LiPF_6 in a 1:1 (v/v, %) solution of ethylene carbonate (EC) and dimethyl carbonate (DMC) was used. The half-cell was in the form of a CR 2032 coin cell. The full-cell was assembled in the form of a pouch. The pouch-type cell was 4.5 cm $\times$ 4.5 cm in size. Al and Ni were used as anode and cathode lead tabs. To obtain enough charging voltage from Si solar cells for LIB charging, a polycrystalline Si solar cell with eight single Si solar cells in a series was purchased from SAVE SOLAR Co. Ltd.

2.2. Characterizations

Large and small particles of NCM622 precursor were prepared using a co-precipitation method. First, large and small particles of NCM cathode were mixed at a 7:3 ratio. Graphite powder was purchased from Sigma Aldrich Co. Ltd. Surface and shape morphologies of NCM as a cathode and graphite as an anode powder were characterized by FE-SEM (model SU-70, Hitachi High-Tech Corporation, Fukuoka, Japan). The acceleration voltage for the FE-SEM measurement was 10 kV. Structures of NCM and graphite powder were analyzed with an XRD (MAX-2500, RIGAKU in Japan) with $\text{Cu-K}\alpha$ radiation. XRD patterns of the powder were obtained in a 2θ range of 10–80° with a scan rate of 2.0°/min and a step size of 0.05°. Diffraction intensity data for a Rietveld refinement analysis were collected in a step-scan mode with a scanning step of 0.02° and a sampling time of 2 s. Rietveld refinement was performed using a FullProf program to obtain the crystal structure parameters of NCM622. Electrochemical characteristics of NCM and the graphite half-cell and the NCM-graphite full-cell were analyzed using a battery cyclizer (WBCS3000S, WONATECH Co., Ltd., Seoul, Korea). Galvanostatic charge and discharge tests of the NCM-graphite full-cell were carried out over a voltage range of 2.6–4.4 V. Charge and

discharge analyses were performed according to various C-rates of the NCM-graphite full-cell. Cycle retention of the NCM-graphite full-cell was examined after 50 cycles. The fill factor, the open circuit voltage, and the short circuit photocurrent density were measured by irradiating 100 mW/cm^2 of light on silicon solar cells with a solar simulator (PEC-L01, Peccell Technologies, Yokohama, Japan).

2.3. Integrated Si Solar Cells-LIB Device

Si solar cells and LIB were directly connected to fabricate an integrated device. The integrated sample was examined in the glovebox (WGB2200S, Woosung Hivac, Daejeon, Korea) installed in the Future Energy Convergence Core Center (FECC). The LIB was charged by irradiating light on Si solar cells with a solar simulator (PEC-L01, Peccell Technologies, Japan). Before the test, the light intensity was calibrated to 100 mW/cm^2 . Photo-charging for the Li-ion battery using the Si solar cells was performed using a solar simulator. Discharge was then performed with a battery cyclor.

3. Results and Discussion

Figure 1 shows FE-SEM images of NCM and graphite powder. The particle size and the shape of NCM powder are shown in Figure 1a,b. The NCM powder consisted of large and small particles with diameters of about $10 \mu\text{m}$ and $3 \mu\text{m}$, respectively. Large particles and small particles were mixed as a bimodal. By mixing large and small particles with optimized ratios, a better energy density could be realized [18]. Park et al. [19] have reported that optimized bimodal-sized active materials can minimize voids and maximize the packing density. A bimodal structure could also improve electrochemical performance due to a large contact area between large particles and small particles [19]. The particle size and the shape of the graphite powder are shown in Figure 1c,d. FE-SEM magnitudes of Figure 1c,d were 1000 and 3000, respectively. The average size of the graphite powder was about $20 \mu\text{m}$. As shown in Figure 1, the size of the graphite powder was greater than that of the NCM powder. Surface morphologies of the graphite powder were smoother than those of the NCM powder.

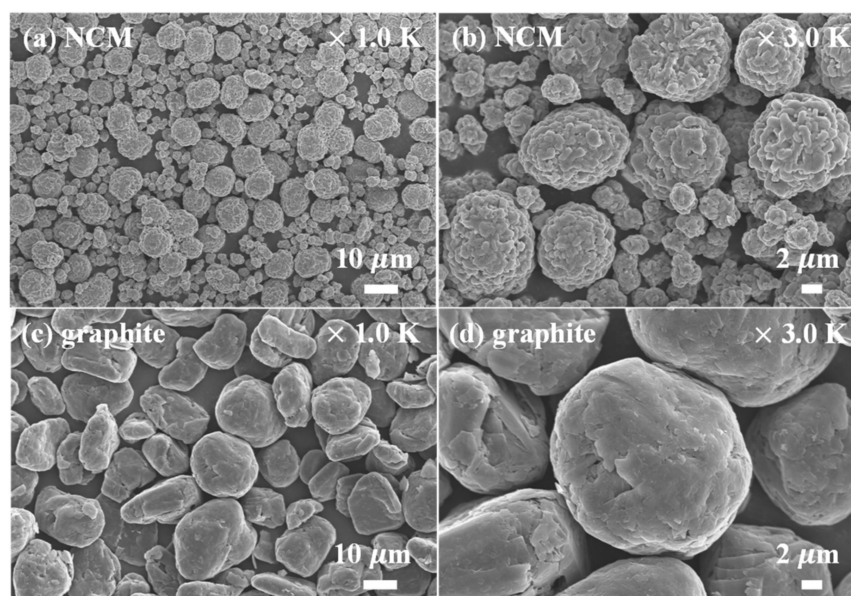


Figure 1. FE-SEM images of (a,b) NCM powder and (c,d) graphite powder.

XRD patterns of NCM and graphite powders are shown in Figure 2a,b. As shown in Figure 2a,b, XRD patterns of NCM and graphite powders and their joint committee on powder diffraction standard (JCPDS) data were compared. It could be noted that XRD patterns of NCM and graphite powders were very close to those of PDF data. Other XRD

patterns related to impurities were not found. Results of XRD patterns confirmed that the NCM cathode powder and the graphite anode powder were successfully synthesized. For detailed structural analysis, Rietveld refinement of the NCM cathode was performed. The results are shown in Figure 2c. The structural parameters of NCM622 obtained from the Rietveld refinement are listed in Table 1. The NCM622 sample presented a hexagonal α -NaFeO₂ structure with an R3m space group. Structural lattice parameters of the NCM 622 cathode are also shown in Table 1. These parameters indicated that the NCM 622 cathode material was well-ordered with a layer structure. The values of c/a and I_{003}/I_{104} are important factors because these values indicate the degree of structural orderliness for layered structural materials. As shown in Table 1, the values of c/a and I_{003}/I_{104} were larger than 4.9 and 1.2, respectively. Thus, the sample had a well-ordered layered structure and a lower amount of Li⁺/Ni²⁺ mixing [20]. The refined atomic parameters of Ni, Co, and Mn were 0.65, 0.15, and 0.20, respectively, as shown in Table 2. While the Li1 was located in the interslab layer, the Ni1, Co1, and Mn1 were in the NCM interlayer. A slight cation mixing between Li and Ni atoms was allowed by constraining the total amount of each element to 1.00 and 0.65, respectively.

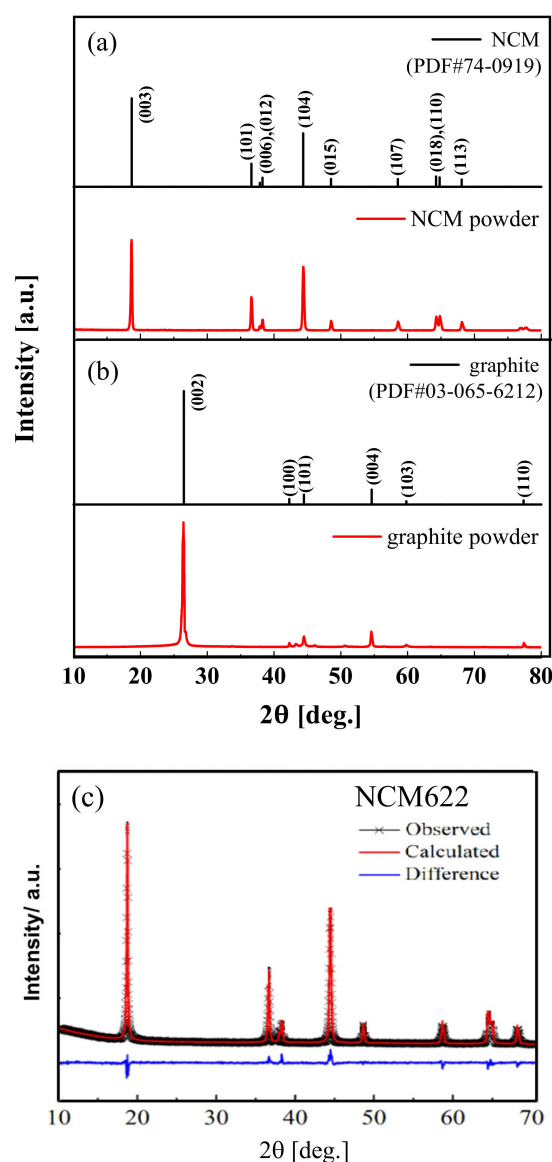


Figure 2. XRD patterns of (a) NCM (JCPDS, 74-0919) powder, (b) graphite (JCPDS, 03-065-6212) powder, and (c) Rietveld refinement of NCM622 powder.

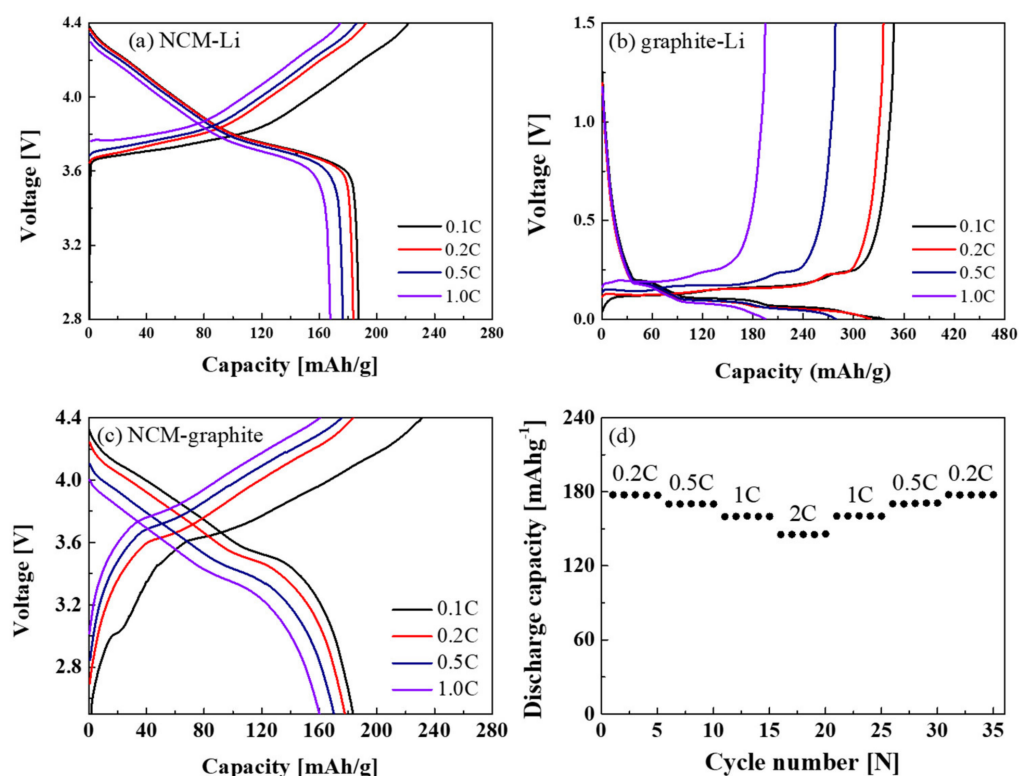
Table 1. Structural parameters obtained from Rietveld refinement of XRD pattern of NCM622.

Sample	<i>a</i> (Å)	<i>c</i> (Å)	<i>c/a</i>	Unit volume (Å ³)	<i>I</i> _{(003)/(104)}
NCM622	2.910	14.292	4.911	101.67	1.575

Table 2. Atomic parameters of the pristine NMC622 material obtained from Rietveld refinement.

Atom	X	Y	Z	Occupancy
Li1	0.00000	0.00000	0.00000	1.0 (fixed)
Ni1	0.00000	0.00000	0.50000	0.65 (fixed)
Co1	0.00000	0.00000	0.50000	0.15 (fixed)
Mn1	0.00000	0.00000	0.50000	0.20 (fixed)
O1	0.00000	0.00000	0.24224 (10)	1.0 (fixed)

The electrochemical results of the half-cell and the full-cell batteries are shown in Figure 3. As shown in Figure 3a, charge and discharge capacities were obtained at 0.1 C, 0.2 C, 0.5 C, and 1 C of the NCM half-cell. The NCM half-cell had a discharge capacity of 187.36 mAhg^{−1} at 0.1 C. The discharge capacity was similar to that reported previously [21]. Figure 3b shows the charge and discharge capacities at 0.1 C, 0.2 C, 0.5 C, and 1 C of the graphite half-cell. The graphite half-cell had a discharge capacity of 336.82 mAhg^{−1} at 0.1 C. Figure 3c shows the charge and discharge capacities at 0.1 C, 0.2 C, 0.5 C, and 1 C of the NCM-graphite full-cell. The NCM-graphite full-cell had a discharge capacity of 183.39 mAhg^{−1} at 0.1 C. The discharge capacity of the full-cell was very similar to that of the NCM half-cell. Figure 3d shows the discharge capacities of the NCM-graphite battery at various C-rates. As shown in Figure 3d, the discharge capacity was almost the same at each C-rate. Thus, the NCM-graphite full-cell had a good electrochemical stability.

**Figure 3.** Electrochemical properties of (a) NCM-Li, (b) graphite-Li, (c) NCM-graphite full cell, and (d) rate capability of NCM-graphite full cell analyzed at various C-rates.

Charge and discharge data of the NCM-graphite full-cell until 50 cycles at the 1 C rate are shown in Figure 4a. The discharge capacity was 160.22 mAhg^{-1} at the 1st cycle, 158.33 mAhg^{-1} at the 5th cycle, 157.98 mAhg^{-1} at the 25th cycle, and 157.41 mAhg^{-1} at the 50th cycle. The NCM-graphite full-cell had a cycle retention of 98.25% after 50 cycles. This suggests that the NCM-graphite full-cell has an excellent lifecycle performance. The NCM-graphite full-cell might exhibit good performance during operation of the integrated device with Si solar cells. The discharge capacity and the Coulombic efficiency for each cycle are shown in Figure 4b. The Coulombic efficiency was 99.68% at the 1st cycle, 99.66% at the 5th cycle, 99.52% at the 25th cycle, and 99.25% at the 50th cycle. Coulombic efficiency was almost constant over 50 cycles.

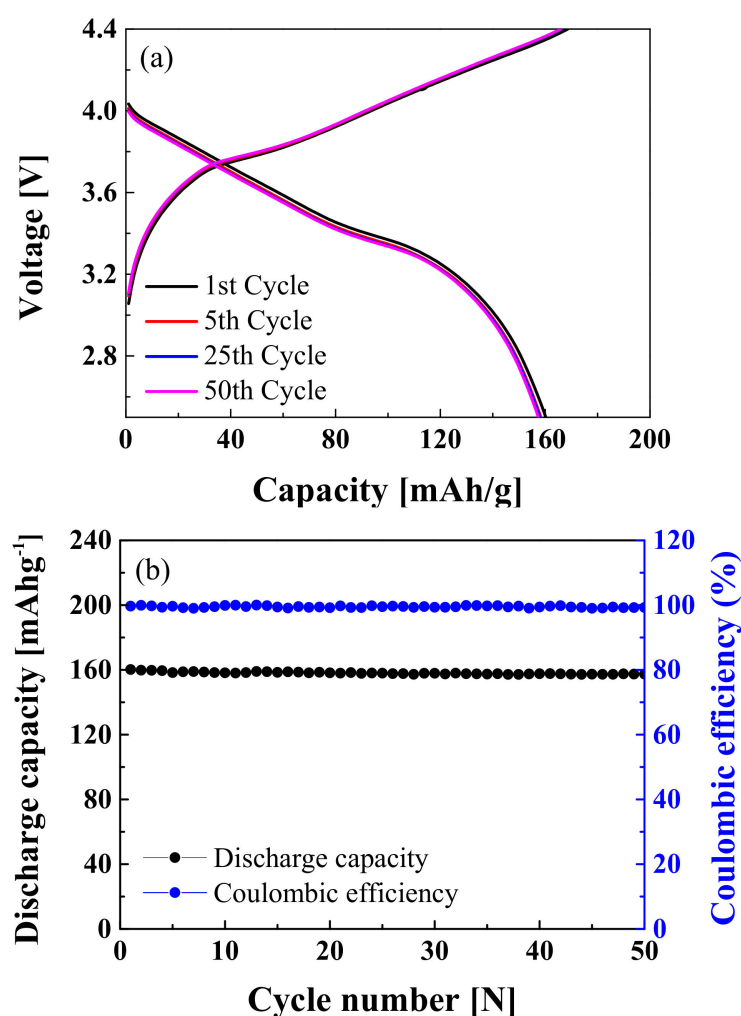


Figure 4. (a) Cycle performance of the NCM-graphite full cell over 50 cycles; (b) discharge capacity and Coulombic efficiency of the NCM-graphite full cell over 50 cycles.

Figure 5 shows a schematic of the integrated device using Si solar cells and the NCM-graphite battery. The voltage range of the NCM-graphite full-cell was 2.6–4.4 V. Therefore, Si solar cells were designed to obtain enough charging voltage for the battery that eight single Si solar cells (0.58 V per each single solar cell) were connected in series. Si solar cells and the NCM-graphite full-cell were directly connected without a power converter. As shown in Figure 5, the integrated Si solar cells and the battery device could be operated by self-photo-charging to the battery. The simple structure without any power converter could be an advantage.

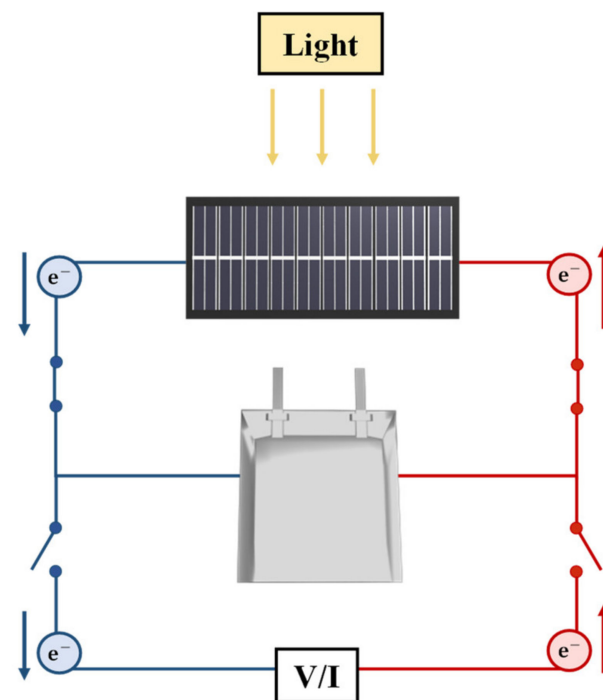


Figure 5. Schematic diagram of NCM-graphite integrated device with Si solar cells.

To examine the electrical property of Si solar cells, current density–voltage (J–V) curves of Si solar cells were characterized. The results are shown in Figure 6. These Si solar cells had a short-circuit photocurrent density of 4.124 mA/cm^2 and an open-circuit voltage of 4.5 V . These Si solar cells had enough voltage to charge the NCM-graphite battery. They had revealed a fill factor of 0.78 and a power conversion efficiency of 14.45% . Generally, efficiencies of commercialized Si solar cells are about $12\text{--}14\%$ [22]. Thus, Si solar cells used in this study had a moderate efficiency of 14.45% based on the literature.

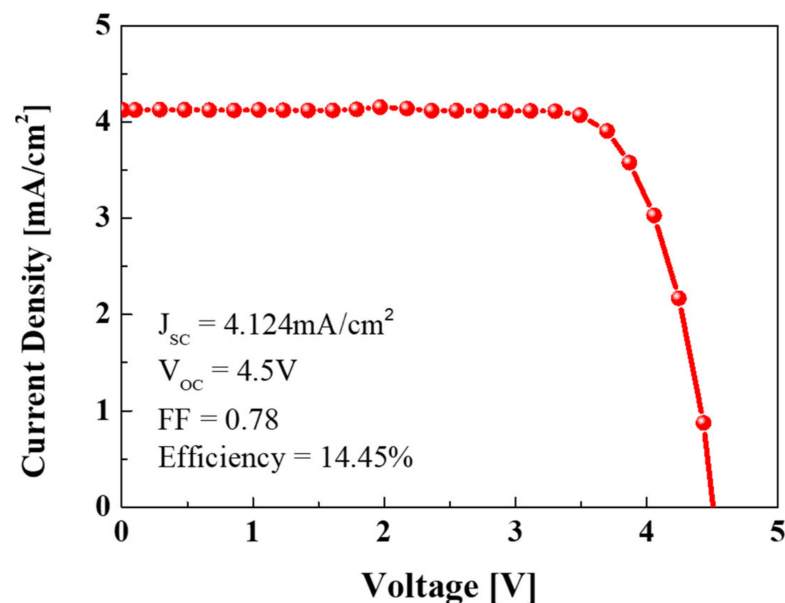


Figure 6. J–V curve of Si solar cells.

Figure 7 shows the results of charging and discharging tests by directly connecting Si solar cells with the NCM-graphite full-cell. The charge–discharge test was performed at a

voltage range of 2.6–4.4 V. The LIB of the integrated device was charged by Si solar cells using a solar simulator. The battery was then discharged using a galvanostatic method over 10 cycles. During charge–discharge over 10 cycles, the charge–discharge behavior was almost the same. This means that the integrated device can be operated stably. The power conversion and storage efficiency of the integrated Si solar cells-NCM-graphite full-cell device was 7.74%. The integrated device showed excellent power conversion and storage efficiency at a high photo-charging voltage [23].

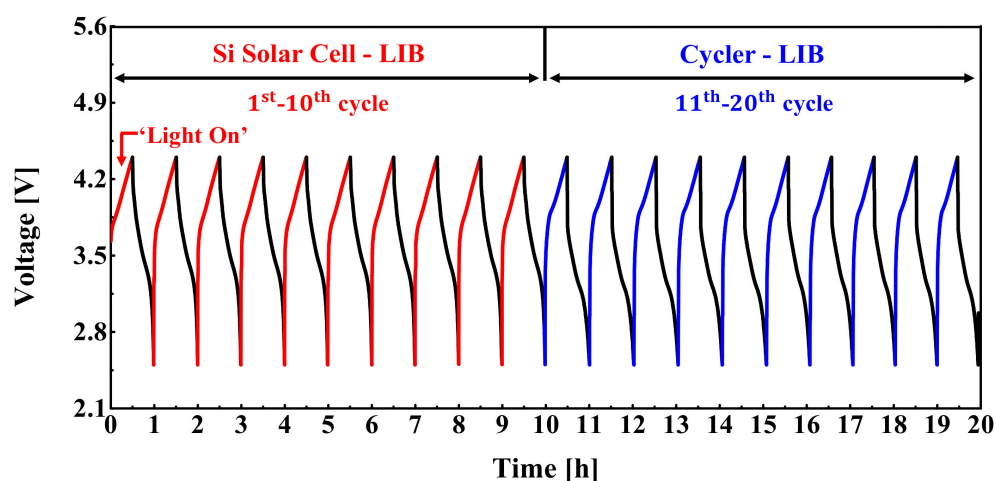


Figure 7. Photo-charging and discharging diagrams of the NCM-graphite full cell using Si-solar cells and charging and discharging with a battery cycler.

In general, LiFePO_4 (LFP) cathode-based batteries (LFP-graphite, LFP-LTO, etc.) are widely used in ESS due to their advantages such as superior thermal stability and chemical stability [24]. These features provide better safety properties than other types of Li-ion batteries. LFP batteries have a low charging voltage. Thus, small amounts of single silicon solar cells were connected to match their charging voltage. LFP batteries are widely used in ESS. However, LFP batteries have lower capacities than NCM batteries [25,26]. NCM batteries have higher charging voltages than LFP batteries. The NCM-graphite full-cell in this study revealed a good cycle life of 98.25% after 50 cycles. Its capacity was higher than the capacity of LFP ($\sim 170 \text{ mAh/g}$). Therefore, the NCM-graphite full-cell could be a promising candidate for an integrated device. The energy storage efficiency from Si solar cells to LIB was 53.56%. After that, the integrated device was charged and discharged for 11–20 cycles with a battery cycler using a galvanostatic power supply in the voltage range of 2.6–4.4 V, similar to the charge–discharge voltage profile of the NCM-graphite battery. Photo-charging of integrated devices is an efficient way of charging using a battery cycler. Therefore, an integrated device of a LIB and a Si solar cell is a promising power system for portable devices, wearable devices, ESS, and EVs.

Figure 8a shows J–V curves of Si solar cells connected to an NCM-graphite full-cell before and after various cycle tests. The current density of Si solar cells connected to the NCM-graphite full-cell was 4.124 mA/cm^2 before the cycle test. As the number of cycles increased, the current density of Si solar cells connected to the NCM-graphite full-cell decreased slightly. After 1, 5, and 10 cycles, current densities were 4.122 mA/cm^2 , 4.114 mA/cm^2 , and 4.099 mA/cm^2 , respectively. These Si solar cells integrated to the NCM-graphite battery revealed an open circuit voltage of 4.49 V after 10 cycles. This open circuit voltage did not change significantly over 10 cycles. Figure 8b shows the fill factor and the energy efficiency of Si solar cells integrated to the NCM-graphite full-cell before and after various cycle tests. The energy efficiency of Si solar cells connected to the NCM-graphite full-cell was 14.45% before the cycle test. As the number of cycles increased, the energy efficiency of Si solar cells connected to the NCM-graphite full-cell decreased slightly. Energy efficiencies of Si solar cells connected to the NCM-graphite full-cell after 1,

5, and 10 cycles were 14.41%, 14.32%, and 14.26%, respectively. The fill factor of the Si solar cells connected to the NCM-graphite full-cell was 0.78 before the cycle test. As the number of cycles increased, the fill factor slightly decreased. The fill factor values of the Si solar cells connected to the NCM-graphite battery after 1, 5, and 10 cycles were 0.78, 0.78, and 0.77, respectively. After 10 cycles, the energy-conversion efficiency of solar cells connected to the NCM-graphite full-cell was 98.68% of the efficiency before the cycling test. Therefore, Si solar cells were suitable devices to supply power to the NCM-graphite full-cell.

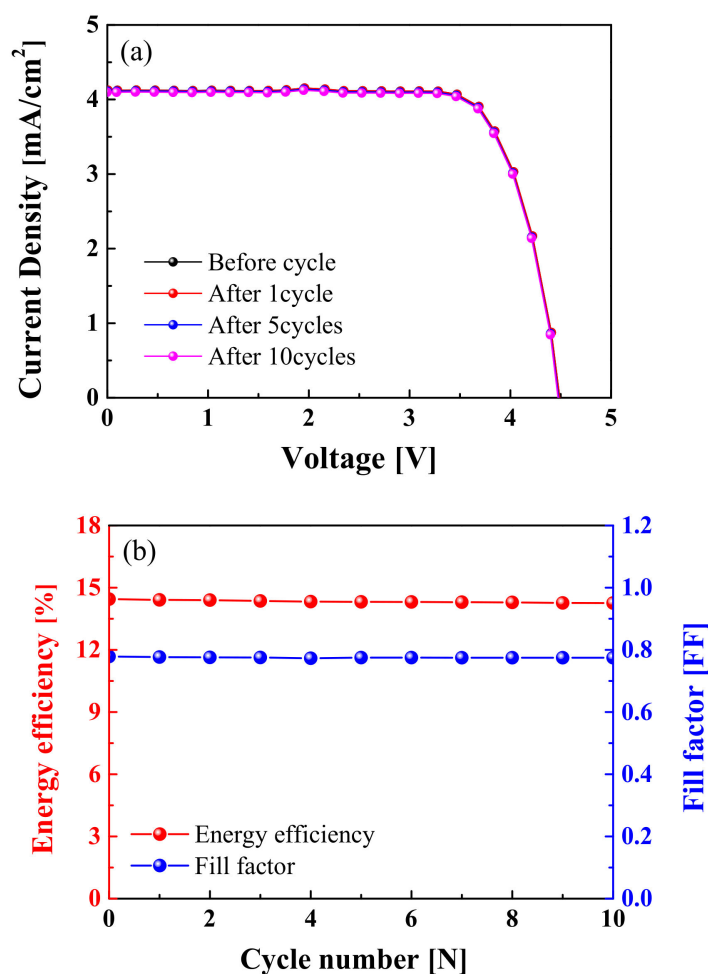


Figure 8. (a) Current density and voltage (J–V) curve; (b) fill factor (FF) and energy efficiency of Si-solar cells integrated to an NCM-graphite battery before and after 10 cycles.

4. Conclusions

In summary, we fabricated a LIB using NCM as the cathode and graphite as the anode. To obtain enough photo-charging voltage, eight single Si solar cells were connected in a series. LIB and Si solar cells were directly connected without any converter. The results of photo-charging with Si solar cells at galvanostatic discharge were very close to charging/discharging with a galvanostatic system. The power conversion efficiency of the NCM-graphite full-cell and Si solar-cells was 7.74%. The energy conversion efficiency of Si solar-cells to LIB was 53.56%, which showed a high energy conversion efficiency.

The cycle retention of the NCM-graphite battery was stable at 98.68% over 50 cycles. In addition, the NCM-graphite full-cell showed a higher capacity than reported LFP-based batteries. Charging of the integrated device could be as effective as charging with a battery cycler. Therefore, an integrated system using an NCM-graphite full-cell is a promising approach to replace an LFP-based integrated system.

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