An Improved High-rate Discharging Performance of “Unbalanced” LiFePO₄ Cathodes with Different LiFePO₄ Loadings by a Grid-patterned Micrometer Size-holed Electrode Structuring

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ABSTRACT

The degradation of charging/discharging capacities in the rate-performance test of lithium iron phosphate (LFP) cathodes with different loading amounts of an active material on both sides of a current collector (i.e., “unbalanced” LFP/LFP cathodes) in a laminated cell (typically composed of anode/separators/unbalanced cathodes/separators/anode) was not observed actually at low C-rates (e.g., 0.1 C). However, the rate-performance data obtained at high C-rates (e.g., >5 C) indicated that the imbalance of the loading amounts of an active cathode material on both sides of an Al current collector causes a significant capacity degradation. We have found that it is possible to prevent the capacity degradation observed at high C-rates by holing the unbalanced LFP/LFP cathodes in a micrometer-sized grid-patterned way (the percentages of the holed area are typically several %) using a pico-second pulsed laser: The non-holed unbalanced LFP/LFP cathodes exhibited a considerable capacity degradation at C-rates which are, for example, larger than 5 C, while the holed ones showed no degradation in capacity even at high C-rates (e.g., 5–20 C). Forming micrometer-sized grid-patterned holes in the LFP/LFP cathodes leads to an improved capacity and high-rate performance of their charging/discharging processes.

Keywords : Lithium Ion Battery, Pico-second Pulsed Laser, C-rate, Cathode
of unbalanced LFP/LFP cathodes and two Li metal anodes in the wide C-rate range of 0.1–20C at different loading ratios and different loading amounts on both sides. The results obtained demonstrate that the high-rate discharging performance can be attained even at 5–20C-rates for the holed (i.e., through-holed and non-through-holed) cathodes in contrast to the non-holed one.

2. Experimental

The non-holed, through-holed and non-through-holed LFP/LFP (LFP: MCC-LR, Mitsubishi Engineering & Shipbuilding Co., Ltd., Japan) cathodes (Fig. 1) in which LFP layers were coated on both sides of an Al current collector foil were prepared using the same electrode materials and conditions as those reported in our previous papers. The preparation is described in detail in the supplementary information (SI). In order to prepare the LFP/LFP cathodes with different loading amounts on both sides of the Al foil, the gap of a doctor-blade coater was suitably adjusted to prepare the LFP layers with different loading amounts, typically ca. 0.7, 1.0, 1.9–2.1, 3.6–4 and 5.6–6 mg cm\(^{-2}\) (Table 1). Holing and characterization of these LFP/LFP cathodes was conducted using a picosecond pulsed laser as described previously \(25-27\) (see 1.2 and 1.3 in the SI). Table 1 lists 23 LFP/LFP cathodes examined in this study and hereafter the individual cathodes will be denoted as No. 1, No. 2 ... and No. 23. The loading amounts, thicknesses and densities of LFP layers on the through-holed (Nos. 1–9), non-through-holed (Nos. 10–14) and non-holed (Nos. 15–23) LFP/LFP cathodes are summarized in Table 1. In numbering the cathodes, as the number increases, the LFP amount located on current collectors decreases. For example, in the through-holed LFP/LFP cathodes of Nos. 1–4, the loading amount is fixed to 5.6–6 mg cm\(^{-2}\) on one-side of a current collector and the loading amount on the another side is 5.6–6, 3.6–4, 1.9–2.1 and 0.7–1.0 mg cm\(^{-2}\), respectively, in Nos. 1, 2, 3 and 4. In the cathodes of Nos. 5–7, the loading amount on one-side of a current collector is fixed to 3.6–4 mg cm\(^{-2}\) and the loading amount on the another side is changed as 3.6–4, 1.9–2.1 and 0.7–1.0 mg cm\(^{-2}\), respectively, in Nos. 5, 6 and 7. Also in the non-through-holed (Nos. 10–14) and non-holed (Nos. 15–23) LFP/LFP cathodes, their numbering was done in the same way. The cathode samples Nos. 1–23 are also recognized by their colored cross-sectional diagrams as shown in Table 1 where the loading amounts of ca. 0.7–1.0, 1.9–2.1, 3.6–4 and 5.6–6 mg cm\(^{-2}\) are represented by color, i.e., yellow, green, blue and red, while considering the LFP loading amounts.

The rate performance of the LFP/LFP cathodes prepared was examined with the cells composed of one of these cathodes and two Li metal anodes, as typically shown for the through-holed LFP/LFP cathodes in Fig. 2, in which the prepared through-holed LFP cathode was inserted between two lithium (Li) metal sheet anodes and separator sheets (HiporeTM, thickness: 25 µm, Asahi Kasei Co., Japan) were also inserted between each side of the cathode and the Li anode. In addition, instead of Li metal sheets, non-through-holed graphite (SMG-N-HE2-20, Hitachi Chemical Company, Ltd., Japan) anodes were used to prepare full cells composed of LFP/LFP cathode and two graphite anodes. Note that the cell system of Fig. 2 is composed of two parallel-connected batteries.

3. Results and Discussion

3.1 Characterization of holed LFP/LFP cathodes with a variety of loading amounts and ratios on both sides

Figure 3(I) shows typical surface SEM images of through-holed LFP/LFP cathodes (Nos. 1–4) having different LFP loadings (and thus different LFP layer thicknesses). On the front surface to which the laser was irradiated (A), the through-holes having the diameters of 36–40 µm (as (A) in Fig. 1) were arranged in a grid-patterned way. The distance between the centers of the closest holes is 167 µm. Around each hole, the crater formed by a mechanical shock of laser irradiation was observed. On the back side from which the laser beam was emitted (B), the formation of such a crater could not be observed. The average crater sizes (\(1\)) and hole diameters (\(2\), \(3\)) of through-holed LFP/LFP cathodes Nos. 1–9 (Fig. 1(A)), and the average crater sizes (\(4\), \(5\), \(6\)) and hole diameters (\(7\), \(8\)) of non-through-holed LFP/LFO cathodes Nos. 10–14 (Fig. 1(B)), which were evaluated from the surface and cross-sectional SEM images of the individual electrodes such as those shown in Fig. 3, are summarized in Table 2. These values obtained from the surface and cross-sectional SEM images agreed well. The cross-sectional SEM images also gave the LFP layer thickness values and thus the density of the LFP layers was calculated to be in the range of 1.4–1.7 g cm\(^{-3}\) (Table 1). It can be seen from Tables 1 and 2 that the crater size (\(1\)) decreased with a decrease in the loading amount (and the thickness) of the LFP layer on the front surface, while the hole diameters of (\(2\) and \(3\)) were not changed largely irrespective of the loading amounts and ratios on both sides. In addition, the opening rates of holes formed on the cathodes (i.e., the percentages of the holed areas) were calculated using the diameters of (\(1\), \(2\), \(3\)) and the results are given in Table 3. In the case of Nos. 1–9, the opening rates for \(2\) and \(3\) were in the range of 3.0–4.5 and 3.2–5.7%, respectively, and were almost the same and independent of the LFP layer thickness, while the opening rate for \(1\) changed largely in the range of 12–24% depending on the LFP layer thickness. As can be seen from Figs. 4(I) and (II) and Tables 1 and 3, the crater sizes (\(4\), \(5\), \(6\)) on the non-through-holed LFP/LFP cathodes were relatively smaller than those on the through-holed ones, probably because of a smaller number of laser shot applied to prepare the non-through-holed cathodes and thus a smaller number of the LFP particles brushed away from the LFP layer surfaces by laser irradiation. A large crack can be observed at the interface of the LFP/LFP layers in the cross-sectional SEM image of sample No. 11. The crack was unfavorably formed in the cross-section formation with a cross-sectional polishing. It is noted here that the laser irradiation is used to form an opening rate. The high-rate performance of the through-holed, non-through-holed and non-holed LFP/LFP cathodes

The high-rate performance of the through-holed, non-through-
holed and non-holed LFP/LFP cathodes prepared was tested at C-rates of 0.1, 0.5, 1, 2, 5, 10, 15 and 20 C. The discharge voltage-discharge capacity curves observed for the through-holed LFP/LFP cathodes Nos. 1–9 are shown in Fig. S1. These curves exhibited the expected behavior depending C-rate, i.e., in the early stage of discharging, DC-IR drop was seen and then the plateau was obtained.

Table 1. Loading amounts of LFP layer on the through-holed (Nos. 1–9), non-through-holed (Nos. 10–14) and non-holed (Nos. 15–23) LFP/LFP cathodes. The samples Nos. 1–23 are presented systematically by their colored cross-sectional diagrams where the loading amounts of ca. 0.7–1.0, 1.9–2.1, 3.6–4 and 5.6–6 mg cm\(^{-2}\) are represented by color, i.e., yellow, green, blue and red, respectively, while considering the LFP loading amounts.

| Nos. | 1 | 2 | 3 | 4 | 5 |
|------|---|---|---|---|---|
| Colored diagrams of cross-sectional LFP/LFP cathodes | | | | | |
| Side of Al current collector | back | front | back | front | back | front | back | front | back | front |
| Loading amount (mg cm\(^{-2}\)) | 5.8 | 5.6 | 5.8 | 4.0 | 6.1 | 2.1 | 5.7 | 1.0 | 3.6 | 3.5 |
| Thickness (µm) | 42 | 45 | 45 | 29 | 45 | 15 | 44 | 7 | 27 | 26 |
| Density of LFP layer (g cm\(^{-3}\)) | 1.6 | 1.4 | 1.5 | 1.6 | 1.6 | 1.5 | 1.7 | 1.6 | 1.6 | 1.6 |

| Nos. | 6 | 7 | 8 | 9 |
|------|---|---|---|---|
| Colored diagrams of cross-sectional LFP/LFP cathodes | | | | |
| Side of Al current collector | back | front | back | front | back | front | back | front |
| Loading amount (mg cm\(^{-2}\)) | 3.8 | 1.9 | 3.9 | 0.9 | 2.1 | 2.0 | 2.1 | 0.7 |
| Thickness (µm) | 28 | 14 | 29 | 7 | 15 | 15 | 15 | 5 |
| Density of LFP layer (g cm\(^{-3}\)) | 1.6 | 1.6 | 1.6 | 1.5 | 1.6 | 1.6 | 1.6 | 1.6 |

| Nos. | 10 | 11 | 12 | 13 | 14 |
|------|----|----|----|----|----|
| Colored diagrams of cross-sectional LFP/LFP cathodes | | | | | |
| Side of Al current collector | back | front | back | front | back | front | back | front | back | front |
| Loading amount (mg cm\(^{-2}\)) | 5.8 | 5.7 | 5.8 | 3.7 | 5.6 | 1.9 | 5.8 | 0.7 | 3.9 | 0.9 |
| Thickness (µm) | 46 | 45 | 44 | 28 | 42 | 15 | 45 | 6 | 29 | 7 |
| Density of LFP layer (g cm\(^{-3}\)) | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.4 | 1.6 | 1.5 | 1.5 |

| Nos. | 15 | 16 | 17 | 18 | 19 |
|------|----|----|----|----|----|
| Colored diagrams of cross-sectional LFP/LFP cathodes | | | | | |
| Side of Al current collector | back | front | back | front | back | front | back | front | back | front |
| Loading amount (mg cm\(^{-2}\)) | 5.8 | 5.7 | 5.8 | 3.7 | 5.6 | 1.9 | 5.6 | 1.0 | 3.6 | 3.7 |
| Thickness (µm) | 45 | 44 | 44 | 30 | 44 | 14 | 41 | 8 | 26 | 27 |
| Density of LFP layer (g cm\(^{-3}\)) | 1.5 | 1.5 | 1.5 | 1.4 | 1.5 | 1.6 | 1.6 | 1.5 | 1.6 | 1.6 |

| Nos. | 20 | 21 | 22 | 23 |
|------|----|----|----|----|
| Colored diagrams of cross-sectional LFP/LFP cathodes | | | | |
| Side of Al current collector | back | front | back | front | back | front | back | front |
| Loading amount (mg cm\(^{-2}\)) | 3.8 | 1.9 | 3.7 | 0.9 | 2.1 | 2.0 | 2.1 | 1.0 |
| Thickness (µm) | 30 | 14 | 30 | 7 | 15 | 15 | 14 | 8 |
| Density of LFP layer (g cm\(^{-3}\)) | 1.5 | 1.6 | 1.4 | 1.5 | 1.6 | 1.6 | 1.7 | 1.4 |

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around 3–3.4 V, and finally the voltage dropped rapidly to 2.0 V. In all the cathodes, the discharge capacities were about 150 and 95 mA h g⁻¹ at 0.1 and 20 C, respectively. In the case of the non-holed LFP/LFP cathodes Nos. 15–23, the high-rate performance (Fig. S2) was significantly different from that obtained for the through-holed ones as mentioned below. Although the discharge capacity at 0.1 C was almost the same for the cathodes examined (around 150 mA h g⁻¹), the discharge capacity at 20 C changed largely depending on the LFP loading amounts and ratios on both sides and the degree of the increase in DC-IR drop with increasing C-rate was much larger than that in the through-holed LFP/LFP cathodes. As can be seen from the comparison of Figs. S1 and S3, the discharge voltage-discharge capacity behaviors were almost the same for the through-holed and non-through-holed cathodes.

The discharge capacity retention vs. C-rate plots obtained based on the above-mentioned results are shown in Figs. 5, 6 and 7 for through-holed, non-through-holed and non-holed LFP/LFP cathodes, respectively, in which the discharge capacity retention was calculated with the following equation.

\[
\text{Discharge capacity retention (\%)} = \frac{\text{the discharge capacity observed at a given C-rate}}{\text{the total theoretical capacity of the cathode used}} \times 100
\]  

(1)

Usually, in the calculation of discharge capacity retention, the discharge capacity observed at 0.1 C is used as a reference capacity. In this case, the total theoretical capacity (5.4 mAh) of the overall unbalanced cathode loading coated on both sides of a current collector was used as a reference capacity because the discharge capacity observed was lower than the theoretical one. The dependence of the discharge capacity retention on the degree of unbalance in through-holed LFP/LFP cathodes is shown in Fig. 5. In Fig. 6, the discharge capacity retention values of the through-holed and non-through-holed LFP/LFP cathodes with the same degree of unbalance of LFP loading amounts, i.e., Nos. 1 and 10, Nos. 2 and 11, Nos. 3 and 12 and Nos. 4 and 13 are compared as a function of C-rate. Figure 7 indicates the C-rate dependence of the discharge capacity retention of the non-holed LFP/LFP cathodes with various degree of unbalance of LFP loading amounts.

The rate performance decreased with increasing C-rate in all cases of these three kinds of the cathodes and at a given C-rate it is in the order of through-holed cathodes (Fig. 5) ≥ non-through-holed cathodes (Fig. 6) ≥ non-holed cathodes (Fig. 7), e.g., at 10 C No. 1 ≥ No. 10 ≥ No. 15 where the loading amounts are 5.6–5.8 mg cm⁻² and the loading ratios µ₁, and No. 3 ≥ No. 12 ≥ No. 17 where the loading amounts are 5.6–6.1 or 1.9–2.1 mg cm⁻² and the loading ratios ≈3. Interestingly, as Figs. 5 and 6 suggest, the high-rate performance of the through-holed and non-through-holed LFP/LFP cathodes was almost the same at a given C-rate irrespective of the LFP loading amounts and ratios on both side of a current collector and the imbalance in LFP loading on both sides did not cause a significant difference in the high-rate performance. On the other hand, in the non-holed LFP/LFP cathodes (Fig. 7), the high-rate performance strongly depended on the degree of the imbalance in LFP loading. For example, No. 18 which has a larger loading imbalance than No. 15 exhibited a larger degradation in the discharge capacity than No. 15, i.e., the discharge capacity retention values of Nos. 15 and 18 were 60 (35) and 18 (0) %, respectively, at 10 (20) C (in contrast to those (100 %) of Nos. 15 and 18 at 0.1 C).

**Figure 2.** Schematic description of the common electronic connection of the cells to the charging/discharging tester and the structure of the cells used for the high-rate performance tests of through-holed, non-through-holed and non-holed LFP/LFP cathodes (typically shown for the cell composed of the through-holed LFP/LFP cathode and Li metal anodes).

**Figure 3.** (I) Typical surface SEM images of through-holed cathodes Nos. 1–4. No. 1 (A), No. 2 (A), No. 3 (A) and No. 4 (A) were viewed from the incident laser plane, and No. 1 (B), No. 2 (B), No. 3 (B) and No. 4 (B) from the laser emission plane. (II) Cross-sectional images of through-holed cathodes Nos. 1–9. The colored cross-sectional diagrams are the same as those in Table 1.
Table 2. LFP/LFP cathodes examined in this study.

| Cathode Nos. | Colored diagrams of cross-sectional LFP/LFP cathodes | Average diameter (µm)¹ | ① | ② | ③ | ④ | ⑤ | ⑥ | ⑦ |
|--------------|-----------------------------------------------------|------------------------|---|---|---|---|---|---|---|
| 1            |                                                     | 92 40 38               | --- | --- | --- | --- | --- | --- | --- |
| 2            |                                                     | 93 37 36               | --- | --- | --- | --- | --- | --- | --- |
| 3            |                                                     | 85 36 42               | --- | --- | --- | --- | --- | --- | --- |
| 4            |                                                     | 81 36 43               | --- | --- | --- | --- | --- | --- | --- |
| 5            |                                                     | 81 35 45               | --- | --- | --- | --- | --- | --- | --- |
| 6            |                                                     | 80 40 45               | --- | --- | --- | --- | --- | --- | --- |
| 7            |                                                     | 79 40 43               | --- | --- | --- | --- | --- | --- | --- |
| 8            |                                                     | 75 35 38               | --- | --- | --- | --- | --- | --- | --- |
| 9            |                                                     | 65 33 34               | --- | --- | --- | --- | --- | --- | --- |
| 10           |                                                     |                        | --- | --- | --- | 56 31 31 61 | --- | --- | --- |
| 11           |                                                     |                        | --- | --- | --- | 56 25 25 37 | --- | --- | --- |
| 12           |                                                     |                        | --- | --- | --- | 54 25 25 30 | --- | --- | --- |
| 13           |                                                     |                        | --- | --- | --- | 52 23 23 25 | --- | --- | --- |
| 14           |                                                     |                        | --- | --- | --- | 50 21 21 45 | --- | --- | --- |
| 15           |                                                     |                        | --- | --- | --- | --- | --- | --- | --- |
| 16           |                                                     |                        | --- | --- | --- | --- | --- | --- | --- |
| 17           |                                                     |                        | --- | --- | --- | --- | --- | --- | --- |
| 18           |                                                     |                        | --- | --- | --- | --- | --- | --- | --- |
| 19           |                                                     |                        | --- | --- | --- | --- | --- | --- | --- |
| 20           |                                                     |                        | --- | --- | --- | --- | --- | --- | --- |
| 21           |                                                     |                        | --- | --- | --- | --- | --- | --- | --- |
| 22           |                                                     |                        | --- | --- | --- | --- | --- | --- | --- |
| 23           |                                                     |                        | --- | --- | --- | --- | --- | --- | --- |

1) Nos. 1–9: through-holed structure, Nos. 10–14: non-through-holed structure and Nos. 15–23: non-holed structure.
2) The colored cross-sectional diagrams are the same as those in Table 1.
3) The holes prepared (①, ②, ... and ⑦ in Fig. 1) do not have a perfect circle shape and thus their average diameters are given.

The similar conclusion can be also obtained in the comparison of the high-rate performance of Nos. 19 and 21. The increase in the surface area of the whole electrode including the sidewalls of holes in through-holed and non-through-holed LFP/LFP cathodes could be calculated to be only 15–20%, i.e., just increasing of the electrode surface area cannot explain the above-mentioned results. Here it should be noted that the cell system (Fig. 2) used for the high-rate discharging performance test is parallel-connected batteries which are composed of two different (or same) LFP/LFP loading cathodes and two Li metal anodes. In this case, different LFP loadings mean different capacities of two batteries. Thus, it may be considered that two batteries would have different internal resistances and thus current through each battery is not the same, being different from the case of parallel-connected batteries with equal LFP/LFP loadings (equal internal resistances) in this case, the current through two batteries must be the same, as being well recognized for the current distribution and discharge behaviors of batteries in series and/or parallel-connected battery packs which are of great concern in battery management systems (BMS).

Table 3. Opening rates of holes on the through-holed (Nos. 1–9) and non-through-holed (Nos. 10–14) LFP/LFP cathodes.

| Cathode Nos. | Average opening rate (%) ¹ |
|--------------|---------------------------|
| 1            | 24 4.5 4.1                |
| 2            | 24 3.8 3.6                |
| 3            | 20 3.6 4.9                |
| 4            | 18 3.6 5.2                |
| 5            | 18 3.4 5.7                |
| 6            | 18 4.5 5.7                |
| 7            | 17 4.5 5.2                |
| 8            | 16 3.4 4.1                |
| 9            | 12 3 3.2                  |
| 10           | 8.8 2.7 2.7 10            |
| 11           | 8.8 1.7 1.7 3.8           |
| 12           | 8.2 1.7 1.7 2.5           |
| 13           | 7.6 1.5 1.5 1.7           |
| 14           | 7.1 1.3 1.3 5.7           |

1) ①, ②, ... and ⑦ show the holes prepared as shown in Fig. 1.
on the thick layer side could be only partially discharged because of its high resistance. On the other hand, the thin layer side could be discharged even at high C-rate. In addition, in the through-holed and non-through-holed cathodes, the access of Li\(^+\) ions through the holes to LFP materials located on their sidewalls is considered to significantly reduce the discharge capacity degradation at high C-rates.

A largely different high-rate discharging performance of the holed (i.e., through-holed and non-through-holed) and non-holed unbalanced LFP/LFP cathodes, typically shown by the comparison of the discharging capacity retention values of Nos. 3, 12 and 17 and Nos. 4, 13 and 18, may be understood by considering the discharging processes on these three kinds of unbalanced cathodes (Fig. 8). In Fig. 8, the discharging processes on Nos. 3, 12 and 17 at 0.1 and 10 C are illustrated schematically. These unbalanced cathodes have the loading ratios of ca. 3, i.e., the LFP loading amounts (and the LFP layer thickness) on both sides of a current collector are ca. 3 times different. From Figs. 5–7, we can see that the discharging capacity retention values of Nos. 3, 12 and 17 are 71 (100), 69 (100) and 20 (100)% at 10 (0.1) C, respectively. At 0.1 C three kinds of the cathodes gave the discharging capacity retention of 100%. At 10 C the discharging capacity retention values of two...
holed cathodes (Nos. 3 and 12) are almost the same (~70), while that of the non-holed one (No. 17) is much smaller (20). This larger discharging capacity retention of the holed cathodes can be considered to reflect the fact that a facilitated transfer of Li\(^+\) ions in the discharging process can occur through both the surface and the sidewalls (produced by forming the holes) of the LFP layer (or LFP/current collector layers), as found recently for the LFP/LFP cathode with the same LFP thickness on both side of a current collector\(^{25-27}\) and the LFP/activated carbon (AC) cathode (in which the thicknesses of LFP and AC layers are different).\(^{28}\)

![Figure 6](image1)

**Figure 6.** Discharge capacity retention vs. C-rate for non-through-holed (●) and through-holed (○) LFP/LFP cathodes. The colored cross-sectional diagrams are the same as those in Table 1.

![Figure 7](image2)

**Figure 7.** Discharge capacity retention vs. C-rate for non-holed LFP/LFP cathodes Nos. 15–23. The colored cross-sectional diagrams are the same as those in Table 1.
In order to confirm the effect of the holing of the unbalanced cathode with different active material loading amounts on both sides of a current collector upon the high-rate discharging performance, three full cells were prepared (Fig. 9), in which the cathodes with the imbalance ratio of ca. 4:1, Nos. 7, 14 or 21, were used as unbalanced cathodes with two non-holed graphite anodes. In these full cells, the loading amounts of graphite particles on two anodes were controlled to make the
capacities of the anode and cathode isolated by a separator sheet unbalanced. The capacity of one (or another) anode was matched with that obtained for the LFP cathode layer which does not face it directly. That is, the capacities of the anode and cathode that do not face each other directly were matched. Usually, if the anode and cathode face each other via a separator sheet and their capacities do not match, the capacity observed corresponds to the minor one of either electrode. In the case of the through-holed unbalanced LFP/LFP cathode (Fig. 9(a)), the discharge capacity retention at 10 C was 70% which was almost equal to that observed for the cell composed of through-holed LFP/LFP cathode and two Li metal anodes (see No. 7 in Fig. 5). On the other hand, the full cells composed of non-through-holed (b) or non-holed (c) unbalanced LFP/LFP cathode and two graphite anodes exhibited a largely decreased discharge capacity. These results demonstrate a significant holing effect of cathode in an unbalanced full cell upon the high-rate discharging performance. Especially, the anode and cathode that do not face each other directly can “face electrochemically” through the micrometer-sized holes in Fig. 9(a), resulting in an improvement in the high-rate discharge performance.

4. Conclusions

In this study, we have fabricated parallel-connected batteries which are composed of two different (or same) LFP/LFP loading cathodes (i.e., unbalanced (or balanced) LFP/LFP cathodes) and two Li metal anodes and examined their high-rate discharging performance. The unbalanced and balanced LFP/LFP cathodes exhibited the same discharge capacities at low C-rate (e.g., 0.1 C), while at high C-rates (e.g., 5–20 C) the former gave a significantly lower discharge capacity than the latter, reflecting the fact that mismatching of internal resistances of two batteries due to different LFP loadings (i.e., different capacities) can lead to the more resistive battery taking a higher current towards the end of the discharging which results in an accelerated capacity fading. This unfavorable discharging performance of the unbalanced LFP/LFP cathodes at high C-rates could be improved using holed cathodes (i.e., through-holed and non-through-holed ones) in which the LFP layer or LFP/current collector layers are holed at a micrometer size in a grid-patterned way. The improvement in the discharging performance at the holed cathodes at high C-rates can be considered to result from a facilitated transfer of Li⁺ ions in the discharging of the thick layer as well as the thin one through both the surface and the sidewalls (produced by forming the holes) of the LFP layer (or the LFP/current collector layers). In order to keep a high discharge capacity at high C-rates even in thick LFP layers, resulting in an improvement of the battery performance of LIBs, the optimization of the diameter of holes formed and the percentage of holed areas (i.e., opening rate) is needed and further work along this line is in progress.

Supporting Information

The Supporting Information is available on the website at DOI: https://doi.org/10.5796/electrochemistry.19-00049.

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