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Supporting Information

Promotional Recyclable Li-ion Batteries by Magnetic Binder with Anti-Vibration and Non-Fatigue Performance

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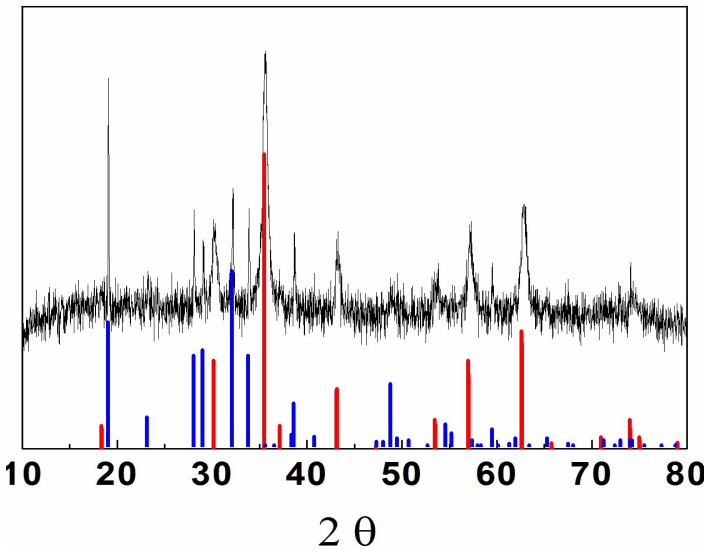
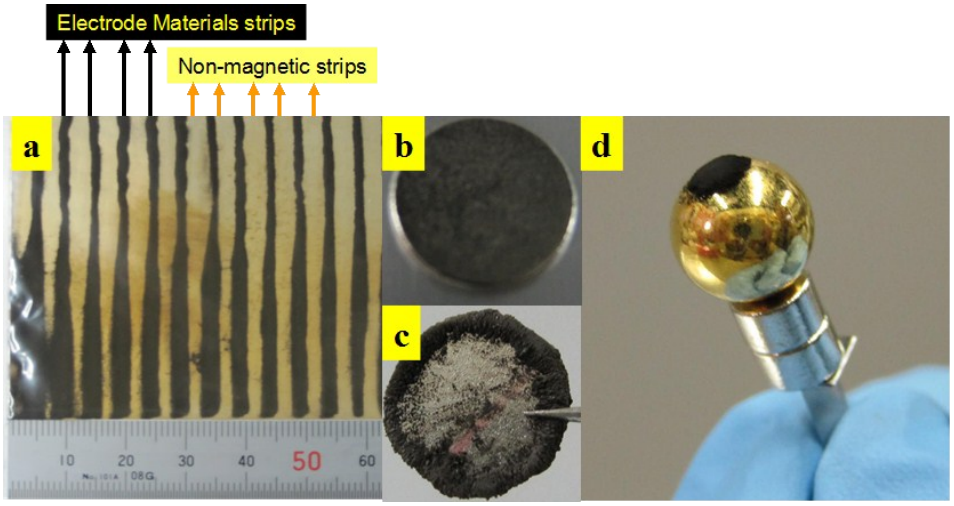


Fig. S1. Phase purity characterization: XRD patterns of the as-synthesized Fe_3O_4. (Red line: JCPDS card of 19-0629 as compound Fe_3O_4, Blue line: JCPDS card of 37-1465 as compound Na_2SO_4).

Materials Characterization: The as-synthesized magnetic Fe_3O_4 have been characterized by XRD and shown in Fig. S1. The patterns can be indexed as compound Fe_3O_4 with JCPDS card 19-0629, and a small amount of impurities can be indexed as Na_2SO_4.


Electrode Materials strips
Non-magnetic strips
a
b
c
d
10
20
30
40
50
60


AIST

Fig. S2. Additional electrode patterns: The distributions of electrode materials, which are manipulated by magnetic field of different magnets. Materials distributions on alternated magnetic and non-magnetic strips, electrode materials formed black line by magnetic field (a); disc magnet with magnetic field in radial directions (b); materials distributions on disc magnet (c); materials distributions on ball magnet with the magnetic poles in the two spherical crowns (d); materials distributed on designed patterns (e).

Materials distribution investigations. The as-synthesized magnetic Fe_3O_4 have been characterized by XRD and shown in Fig. S1. The patterns can be indexed as compound Fe_3O_4 with JCPDS card 19-0629, and a small amount of impurities can be indexed as Na_2SO_4. Materials distributions on the electrode surface can be easily manipulated by designing the magnetic field distribution of MCC. Figure S2 presents the powder distributions on magnets with different field distributions. In Fig. S2a, the electrode materials were adhered on a board with alternated magnetic and non-magnetic strips, which leads to a stripe distribution of the powder. As to the thin disc magnet, the powder was gathered on the edges as shown in Figs. S2b and S2c. In this research, we choose the conductive metal coated spherical magnet as MCC. The magnetic field is strongest on both magnetic poles, so that the powder gathered around the pole of spherical magnet (Fig. S2d). Thereby, the powder distribution can be easily tuned by designing the MCC. Inspired from the magnetic tape, the future MCC can be designed arbitrarily by painting on a magnet slide with a special magnetic pen. It is highly expected of using this design in non-liquid printing batteries and the flexible batteries.

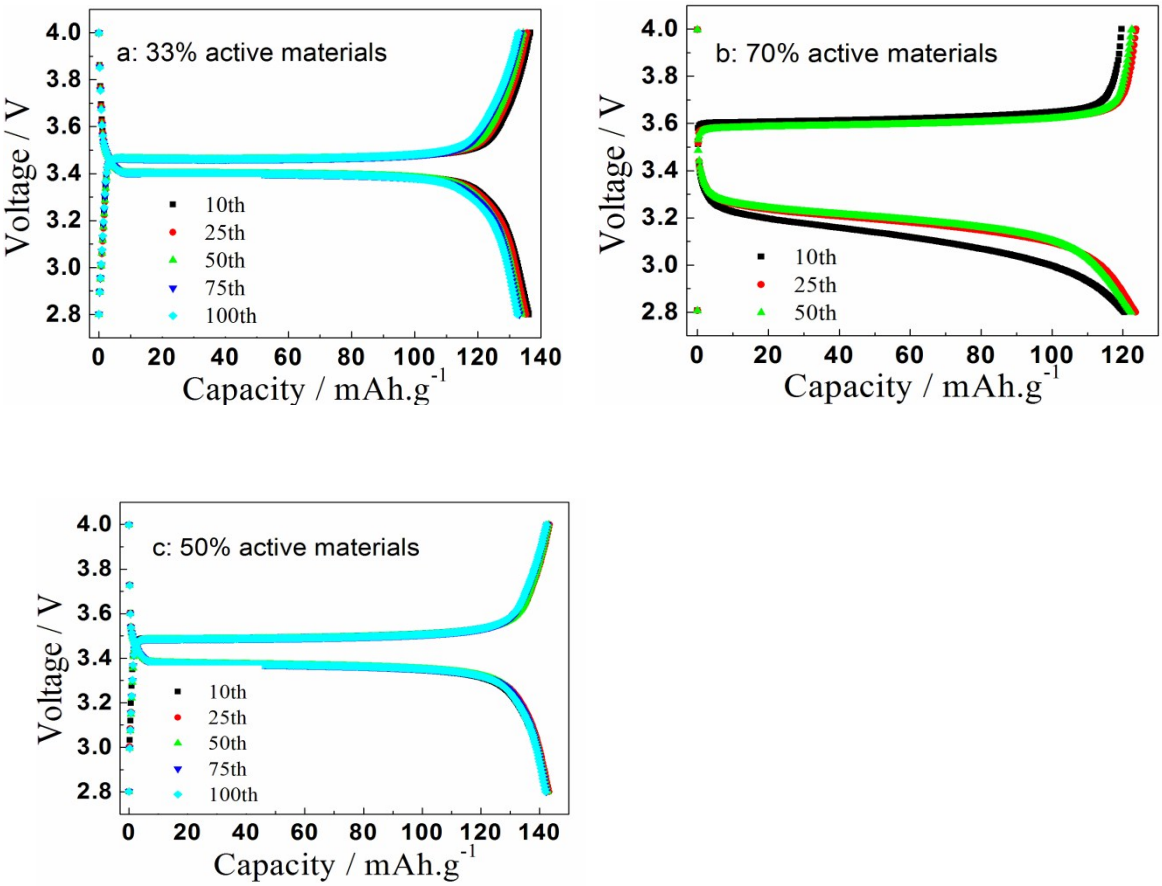


Fig. S3. Additional Voltage profile with different binder contents: Charge-discharge profiles of MME with different magnetic binder amount. The mass ratio of $LiFePO_4$:Fe_3O_4:conductive agent is 1:1:1 (a), 70:15:15(b), and 50:35:15(c).

With improved active material ratio. The magnetic adhesion distinctly impacts the contact between the electrode materials and the current collectors, which is determined by the binder amount and the magnetic susceptibility. Thus, we also investigated the MMEs with different ratios of magnetic binder. Herein, we changed the amount of active material $LiFePO_4$ as 33%, 50% and 70%, and adjusted magnetic binders and conductive agents as well. Galvanostatic cycling tests were conducted in the voltage range of 2.8-4.0 V with a current density of 170 mA/g. Charge-discharge profiles with different materials ratios were shown in Fig. S3. There is scarcely any capacity or voltage decay during the 50 or 100 cycles, which means a non-fatigue property. Compared between Fig. S3a and S3c, there is not any obvious difference for the active material of 33% and 50%. And the excess conductive agent has no evident effect on the electrochemical performance. Further increasing the ratio of $LiFePO_4$ to 70%, there is a serious performance degradation, such as the decreased capacity and the increased polarization. As shown in Fig. S3b, although the samples with 70% active materials showed a stable cycle performance, the discharge capacity reduced to 120 mAh/g. The average charge potential increased from 3.5 V to 3.6 V, and the discharge potential decreased from 3.4 V to about 3.2 V. There was a serious polarization for the high $LiFePO_4$ content. Compared with Fig. S3c, they have the same conductive agent contents, and only the amount of $LiFePO_4$ is increased by decreasing the magnetic Fe_3O_4 binders. Less magnetic binder leads to the poor contact between the electrode material and the MCC. This is the main reason of the degraded electrochemical performance. Thereby, the good contact between the electrode materials and the MCC is vitally important for the battery performance. Furthermore, the saturation magnetization of Fe_3O_4 in this experiment is only 45 emu/g, which is far less than that of 92 emu/g for the bulk materials and could be improved further. And the surface modification of magnetic binder with conductive layer can increase the electronic conductivity and reduce the additional conductive agents. These researches are also under doing in our group now.