nature materials | SUPPLEMENTARY INFORMATION
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Memory effect in a lithium-ion battery

Supplementary Information S1: Enhancing the memory effect by repeating the memory-writing cycle.

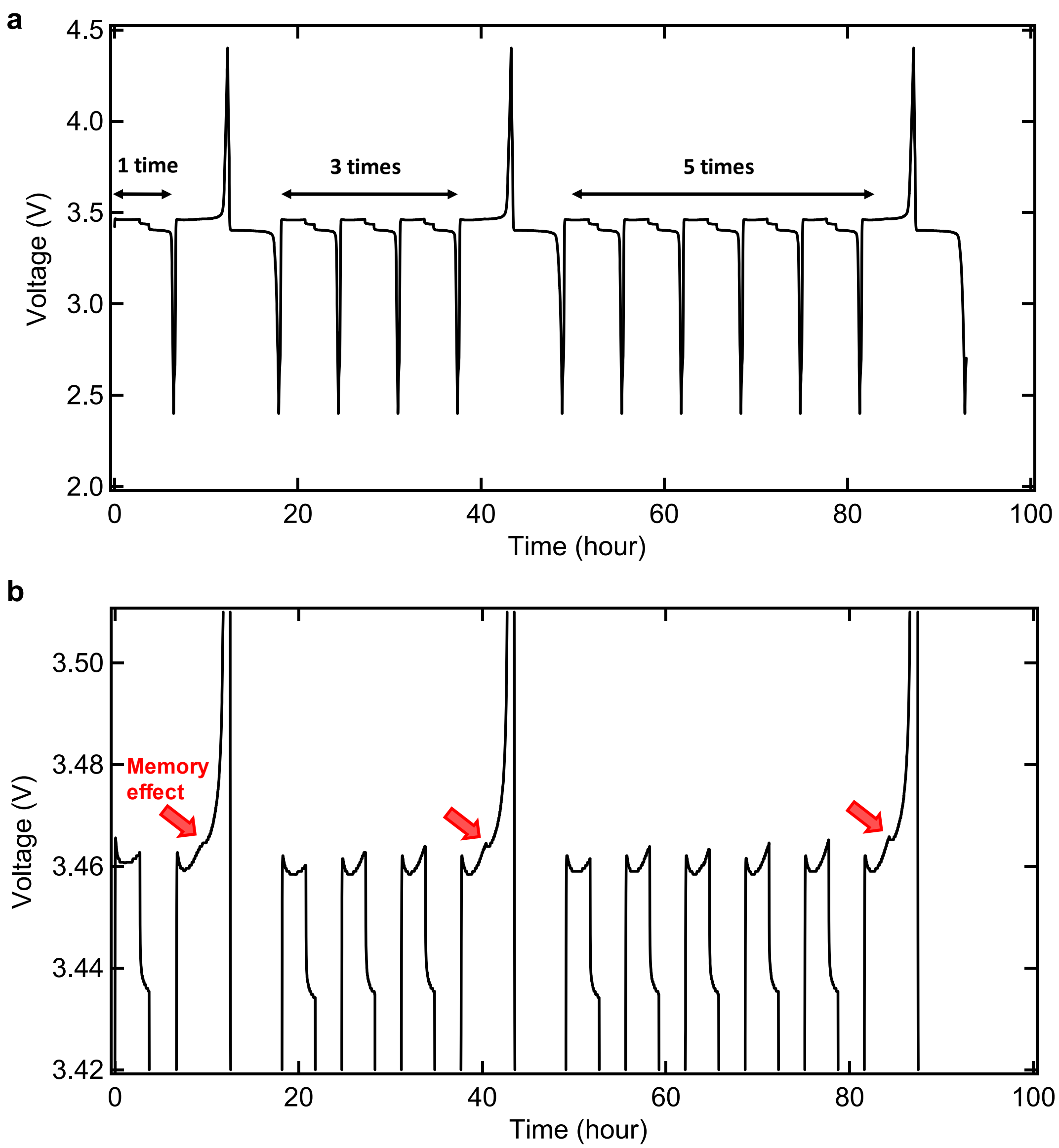

Fig. S1. | Charge and discharge curve of $LiFePO_4$ under the memory-effect conditions with different repetitions of the memory-writing cycle. A sequence of 12

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cycles under three memory-effect conditions; **a**, 1st and 2nd cycle: the memory effect with one memory-writing cycle; 3rd-6th cycle: the memory effect with three memory-writing cycles; 7th-12nd cycle: the memory effect with five memory-writing cycles, between 2.4 and 4.4 V. **b**, Enlarged view of Fig. S1a between 3.42 and 3.51 V.

Supplementary Information S2: The effect of the current rate in the memory releasing cycle.

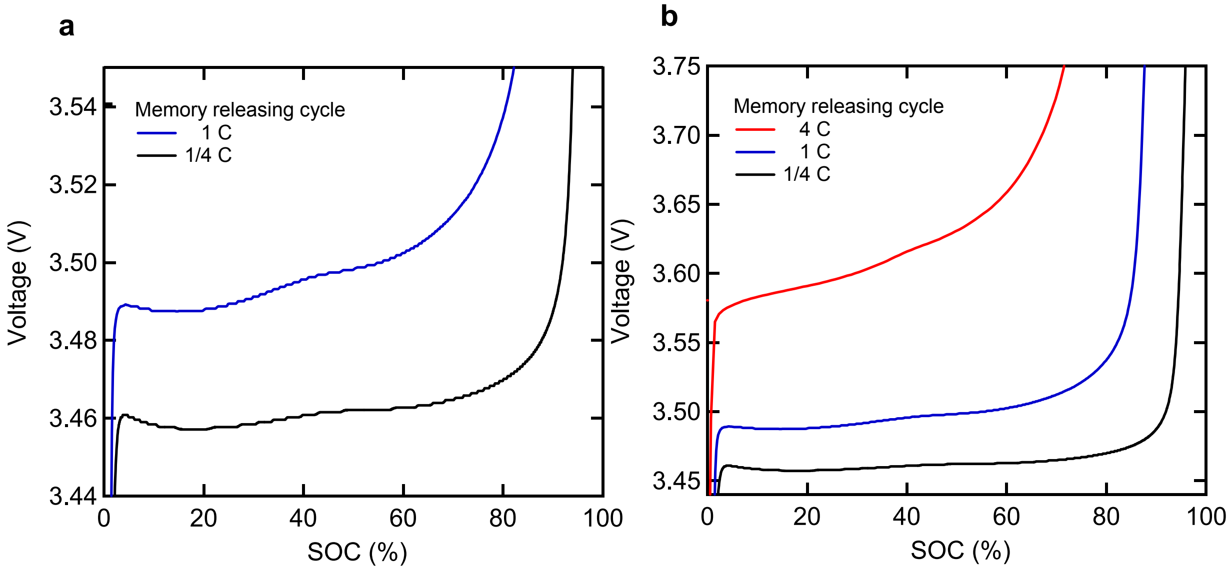

Fig. S2. | Charge curves of the memory-releasing cycle of $LiFePO_4$ at different current rates; a, 1/4 C and 1C rate. **b,** 1/4, 1C, and 4C rate. The conditions of the memory-writing cycles were identical. The depth of the memory-writing cycles was 50% and the current rate was 1/4C. The memory writing cycles had 1 hour rest time between charge and discharge in the open-circuit state. The rest time between the memory-writing cycle and the memory-releasing cycle was 10 minutes.

Supplementary Information S3: Demonstration of the memory effect of $LiFePO_4$ in a three-electrode cell.

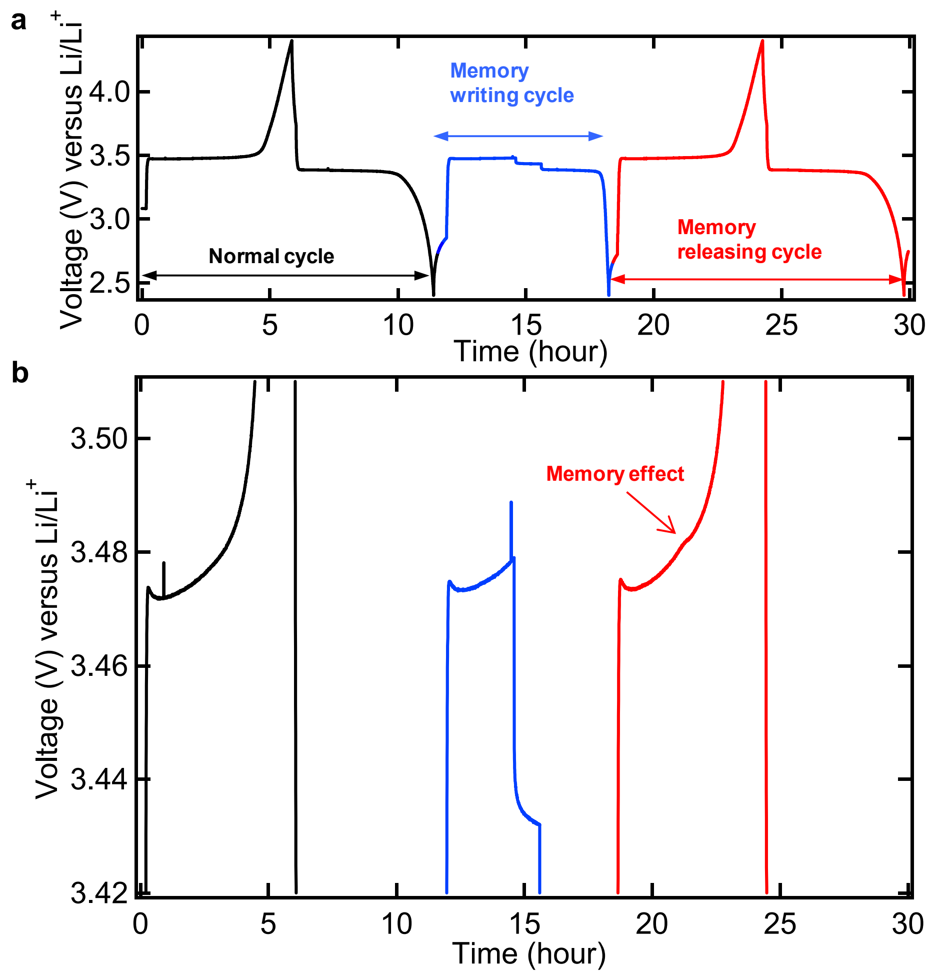

Fig. S3. | Charge and discharge curve of $LiFePO_4$ under 'memory-effect conditions' at a SOC of 50% in a three-electrode cell with a Li-metal reference

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electrode. a, A sequence of three cycles. First cycle (black line): Normal charge and discharge curve between 2.4 and 4.4 V vs. Li/Li^+; second cycle (blue line, memory writing cycle): charge up to a SOC of 50% and full discharge until 2.4 V; third cycle (red line, memory releasing cycle): full charge and discharge between 2.4 and 4.4 V. **b**, Enlarged view of Fig. S3a between 3.42 and 3.51 V.

Supplementary Information S4: Demonstration of the memory effect of $LiFePO_4$ by using a very thin electrode.

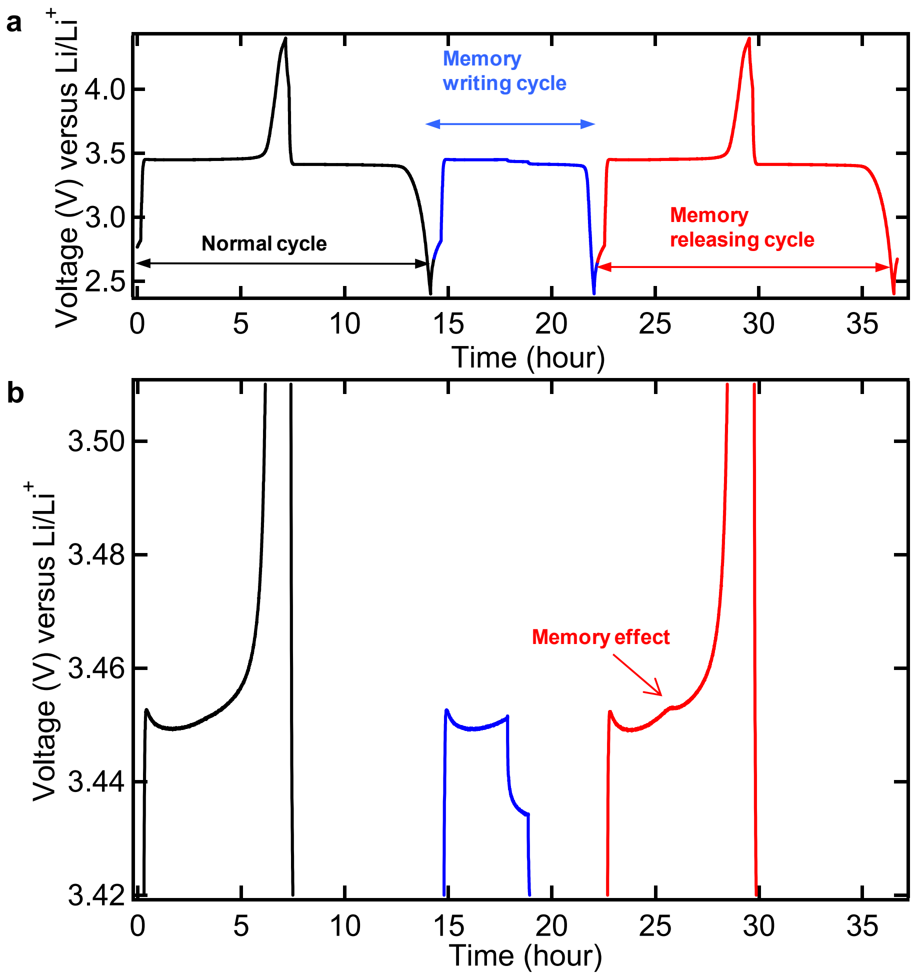

Fig. S4. | Charge and discharge curve of a very thin electrode under 'memory-effect conditions' at a SOC of 50%. The active material loadings of the thin electrodes were 0.14-0.17 mg/cm^2 (standard electrodes in this study: 5 mg/cm^2). **a**, A sequence of three cycles. First cycle (black line): Normal charge and discharge curve; second cycle (blue, memory writing cycle): charge up to a SOC of 50% and full discharge; third cycle (red line, memory releasing cycle): full charge and discharge. **b**, Enlarged view of Fig. S4a between 3.42 and 3.51 V.

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Supplementary Information S5: The effect of the current rate in the memory writing and releasing cycle.

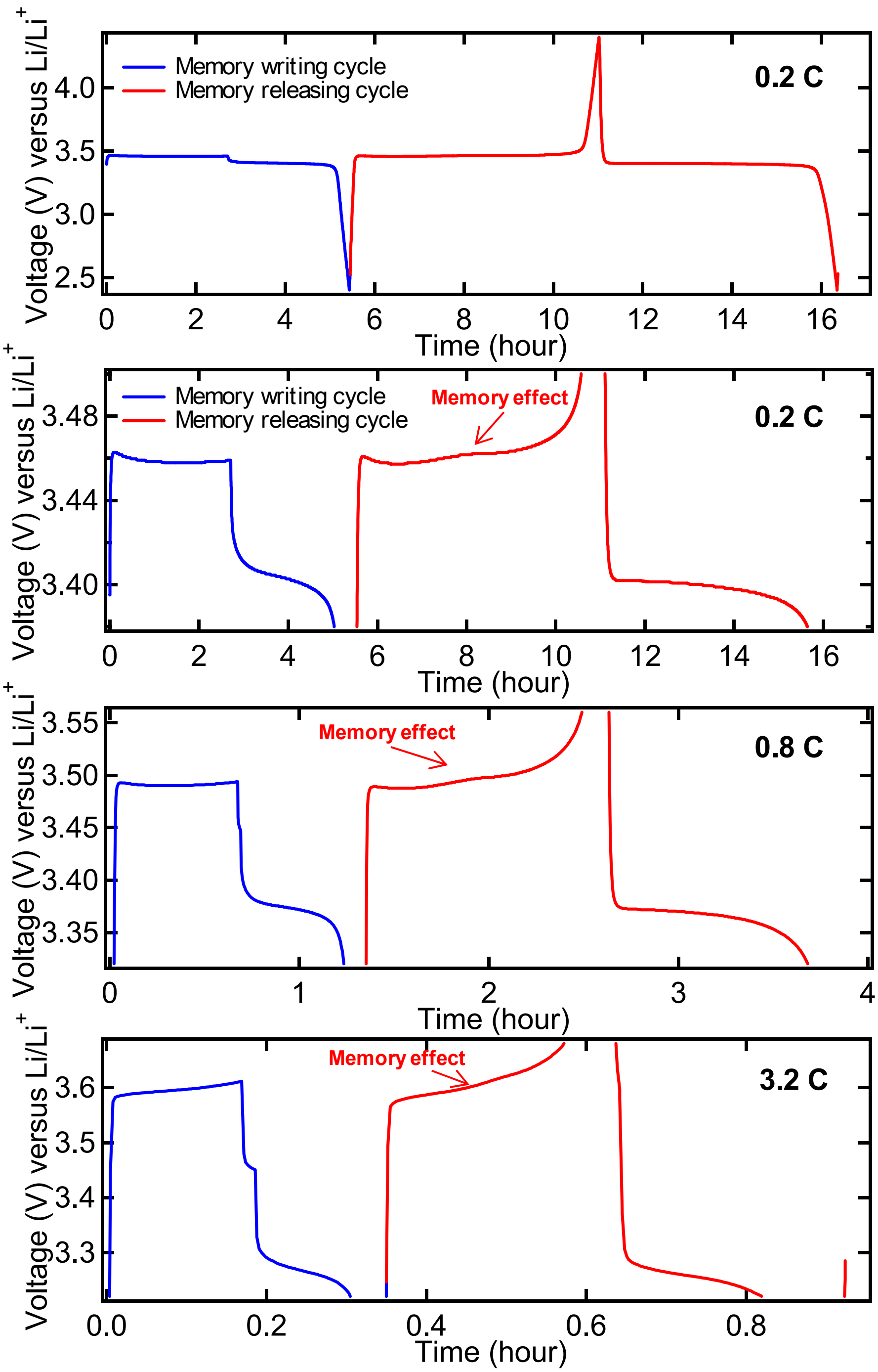

Fig. S5. | Charge and discharge curve of the memory-writing and memory-releasing cycle of $LiFePO_4$ at different current rates; a, First cycle (blue line, memory writing cycle): charge up to a SOC of 50% and full discharge until 2.4 V; second cycle (red line, memory releasing cycle): full charge and discharge between 2.4 and 4.4 V at about 0.2 C rate. **b**, Enlarged view of Fig. S5a between 3.38 and 3.50 V. **c**, Enlarged view between 3.32 and 3.56 V at about 0.8 C rate. **d**, Enlarged view between 3.2 and 3.7 V at about 3.2 C rate. The current rates of the memory-writing and memory-releasing cycles were identical. The memory-writing cycle included a rest time of 1 minute between charge and discharge in the open-circuit state. The rest time between the memory-writing cycle and the memory-releasing cycle was 1 minute.

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Supplementary Information S6: The effect of rest time between the memory writing cycle and the memory releasing cycle

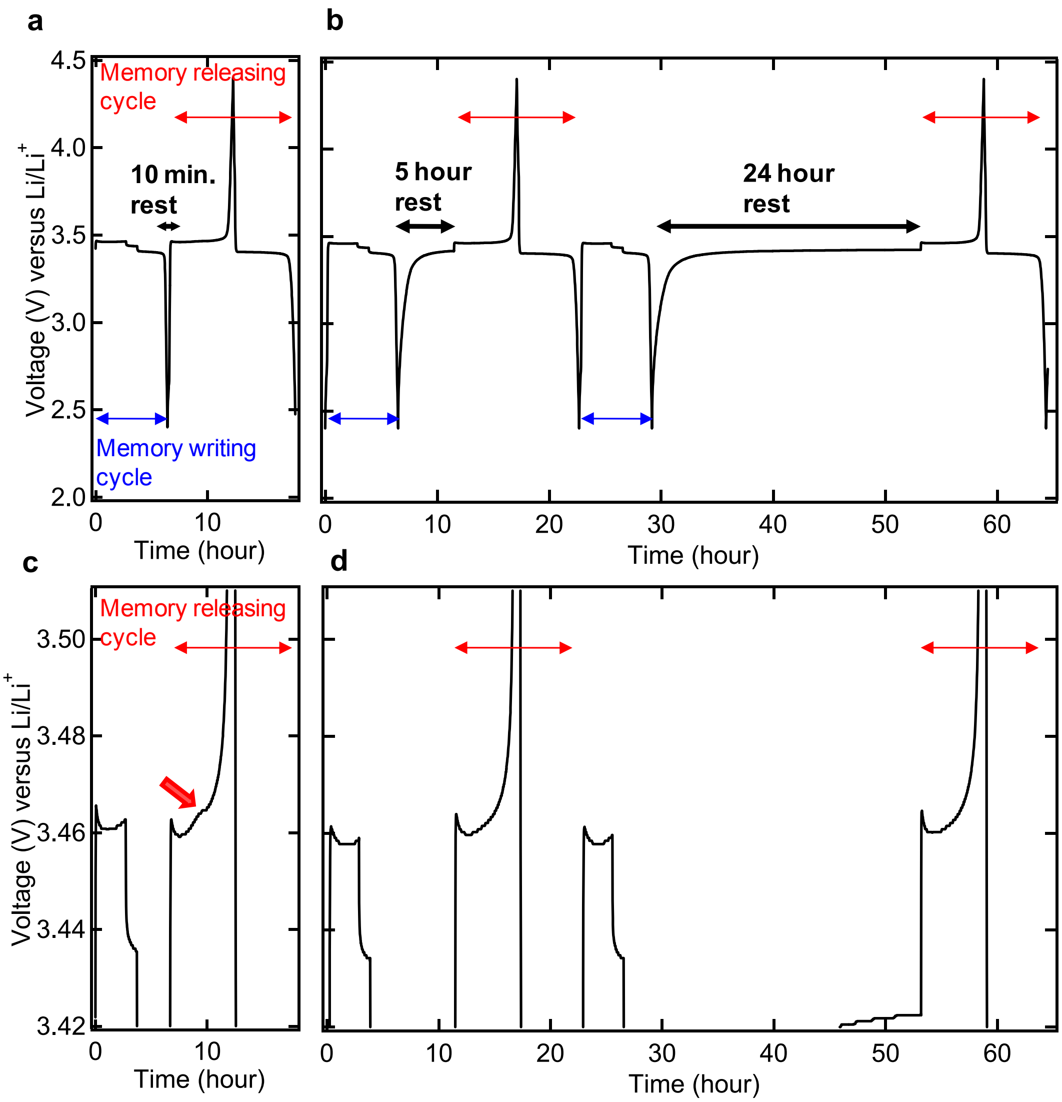

Fig. S6. | Charge and discharge curve of $LiFePO_4$ under the memory effect conditions with different rest time between the memory writing cycle and the memory releasing cycle; a, 10 minute rest time (standard condition in this study) **b**, 5 and 24 hour rest times, **c, d,** Enlarged views of Fig. S6a (c) and S6b (d) between 3.42 and 3.51 V.

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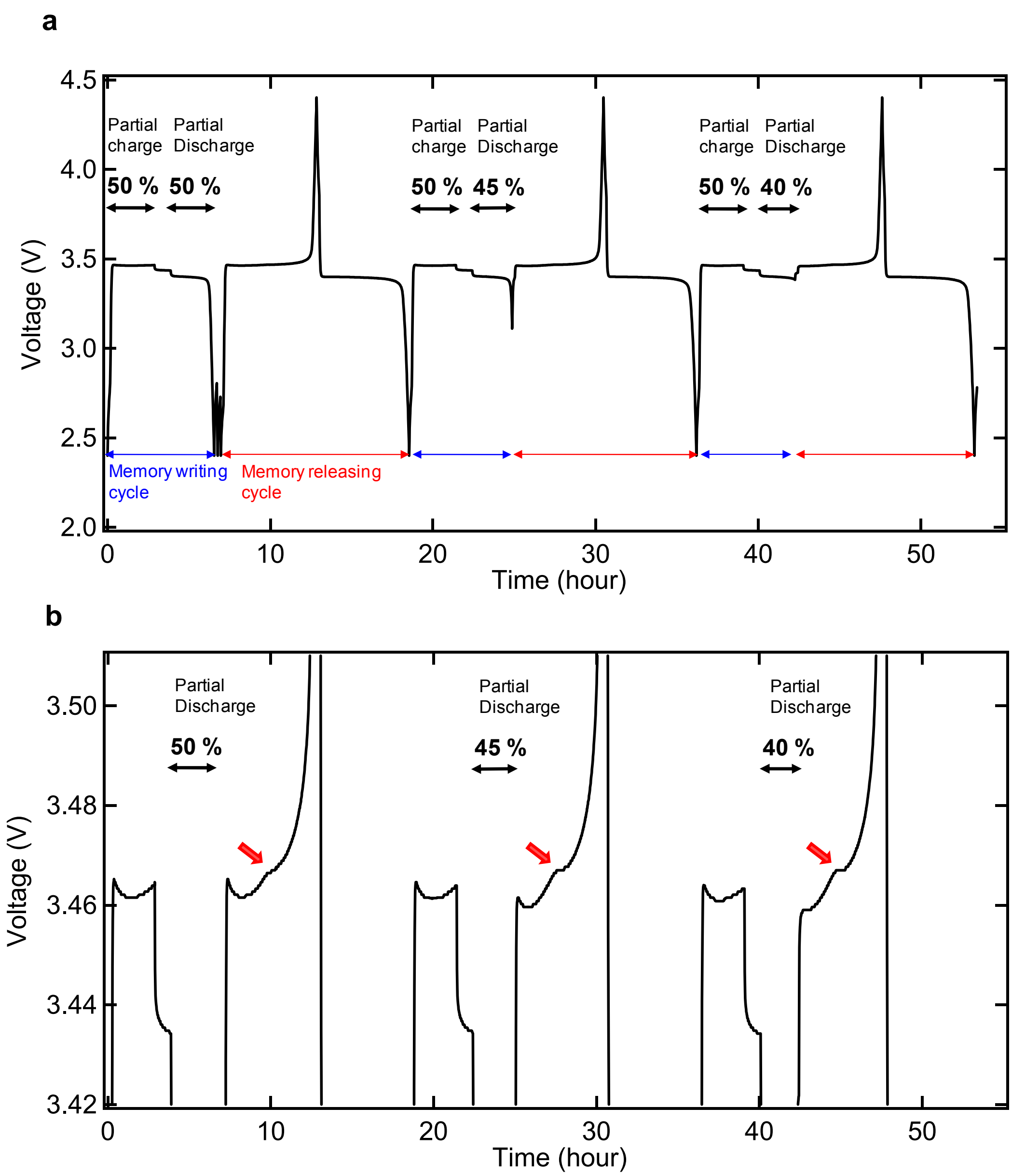
Supplementary Information S7: The effect of the depth of discharge of the memory-writing cycle



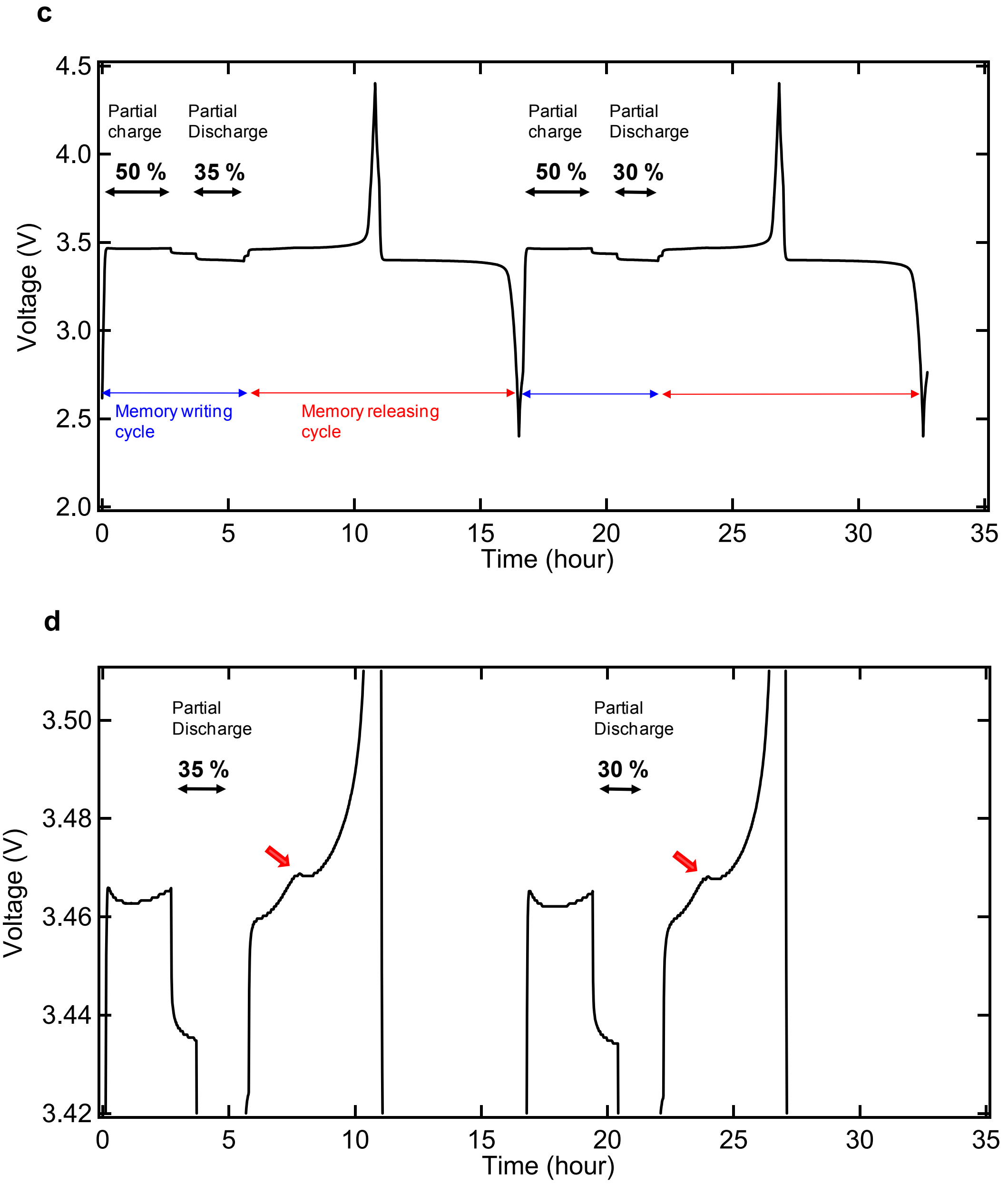


Fig. S7. | Charge and discharge curve of $LiFePO_4$ under the memory effect conditions with different depth of discharge of the memory writing cycle; a, 50% (standard condition in this study), 45%, and 40%, **c,** 35% and 45%. **b, d,** Enlarged views

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of Fig. S7a (b) and S7c (d) between 3.42 and 3.51 V. The rest time between the memory-writing cycle and the memory-releasing cycle was 10 minute in all measurements.

Figure S7b and S7d show significant difference not only in the memory bumps but also in the initial overshoots of the charge in the memory-releasing cycles. With decreasing depth of discharge of the memory writing cycle, the memory bumps were increasing in size; on the other hand, the initial overshoots of the charge in the memory-releasing cycles were decreasing. The latter behaviour also supports our proposed mechanism. According to our mechanism, the initial overshoots come from the first group of Fig. 5f (upper group) and the memory bumps come from the second group (lower group). When the depth of discharge of the memory writing cycle is shallow, a part of the first group still consists of two-phase coexistence particles between Point B and C in Fig. 5. The triggering condition for the overshooting is “Charging from the condition of Fig. 5d”; in other words, “Charging from the condition without two-phase coexistence phase particles”. So, the remaining two-phase coexistence particles will weaken the triggering condition as well as the overshooting. Charging two-phase coexistence particles is easier than charging particles at point A to point B in Fig. 5. The beginning of charge of the memory-releasing cycle after the shallow-depth discharge is led by the remaining two-phase coexistence particles, therefore the overshooting doesn't appear.

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Supplementary Information S8: The effect of long rest time after shallow-depth discharge on the memory-writing cycle

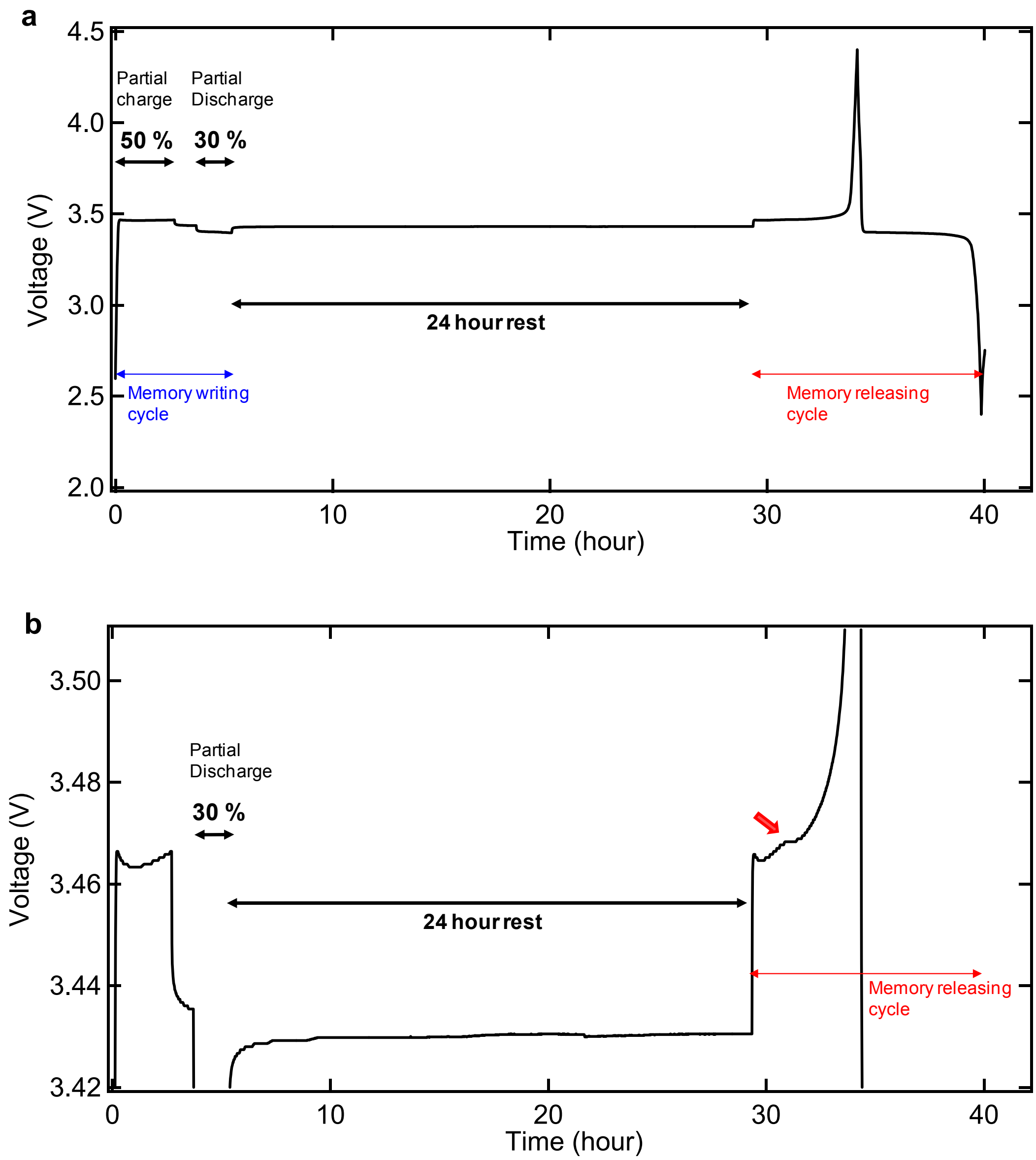

Fig. S8. | The effect of long rest time after shallow-depth discharge on the memory-writing cycle. a, Charge and discharge curve of $LiFePO_4$ under the memory effect condition with 30% depth of discharge of the memory writing cycle and 24 hour rest time between the memory-writing cycle and the memory-releasing cycle**. b,** Enlarged views of Fig. S8a between 3.42 and 3.51 V.

The overshoot behaviours of the memory-releasing cycles between Fig. S7d and Fig. S8b are comparable. Even if the overshooting disappeared after 30% discharge in the memory-writing cycle (see Fig. S7d), the overshooting appeared again after 24 hour rest time, as shown in Fig. S8b. This can be interpreted as long rest times making the remaining two-phase coexistence particles disappear. The remaining two-phase coexistence particles will establish homogeneous Li-poor or Li-rich phases through interactions with other particles during the long rest time, due to the chemical potential gaps. The disappearance of the two-phase coexistence particles lets the initial overshoot of the charge curve appear again.