

## Supplementary Information

### **Role of the Fe-FeCl<sub>2</sub> Contact Interface in Promoting Redox Reversibility and Electrochemical Kinetics in Fe/FeCl<sub>2</sub>-Graphite Molten Salt Battery**

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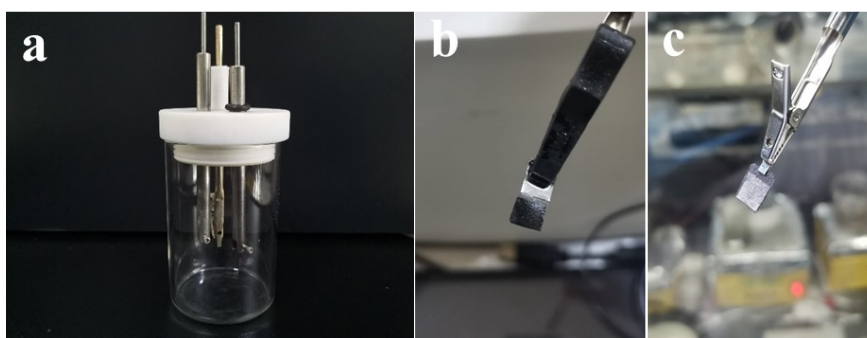
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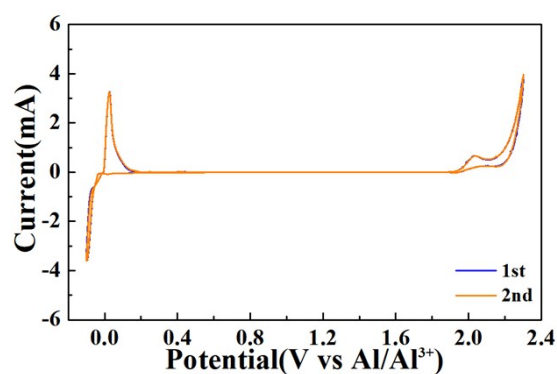
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Figure **S18**. The original chart of the adsorbed energy calculation (-38.934eV) of Fe<sup>0</sup>

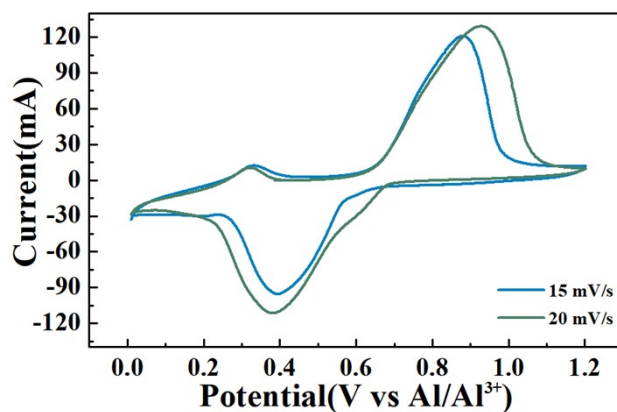
cluster at Fe (211).....S10



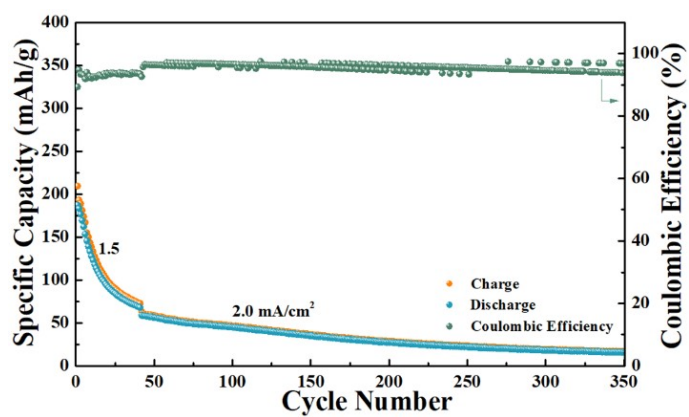
**Figure S1.** (a) Schematic diagram of three-electrode testing device. (b) The  $\text{FeCl}_2\text{-CR}$  electrode prepared by powder calendaring. (c) The graphite electrode clamped by molybdenum mesh.



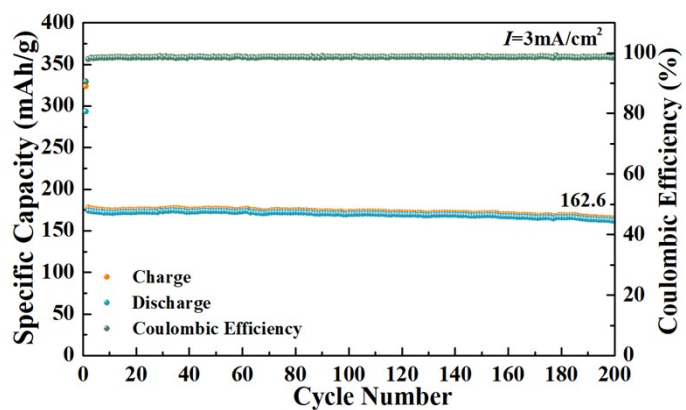
**Figure S2.** CV Curves of the Au electrode in three-electrode system at 10 mV/s.



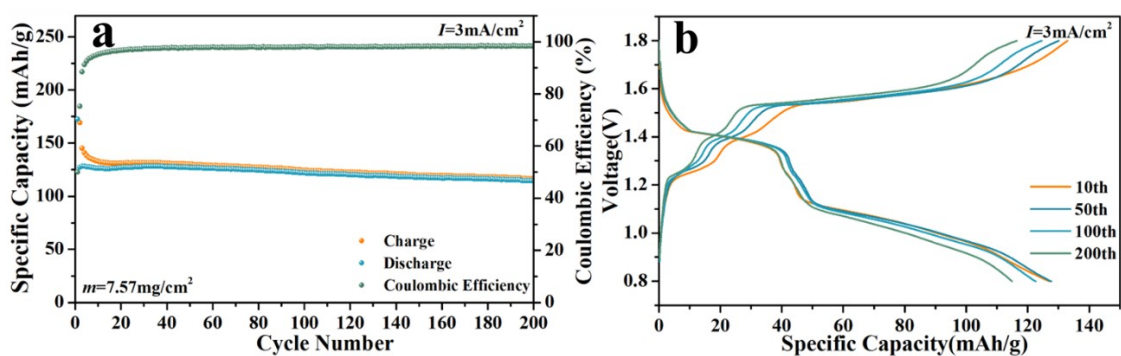
**Figure S3.** CV Curves of the Fe- $\text{FeCl}_2\text{-CR-60}$  electrode in three-electrode system at 15 mV/s and 20 mV/s.



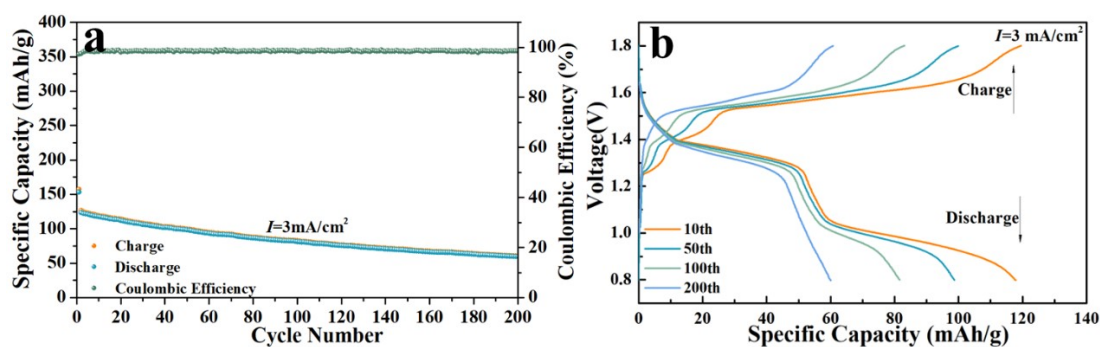
**Figure S4.** Cycling performance of the FeCl<sub>2</sub>-CR electrode in Fe/FeCl<sub>2</sub>-Graphite molten salt battery.



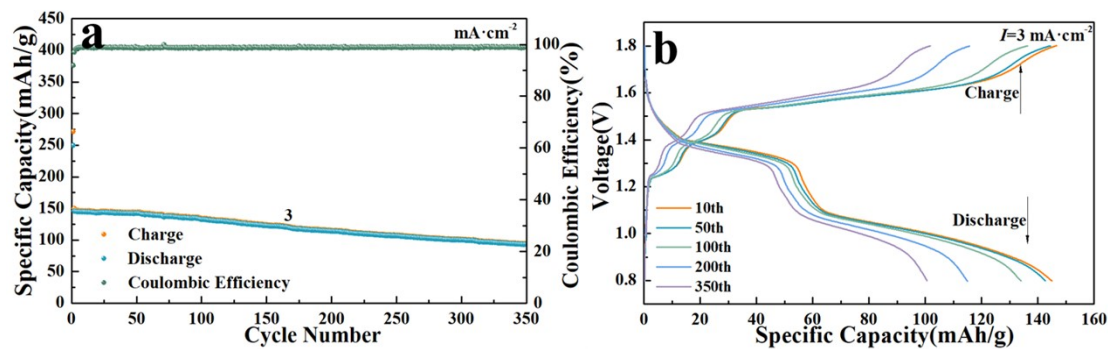
**Figure S5.** Cycling performance of the Fe-FeCl<sub>2</sub>-CR-60 electrode in Fe/FeCl<sub>2</sub>-Graphite molten salt battery.



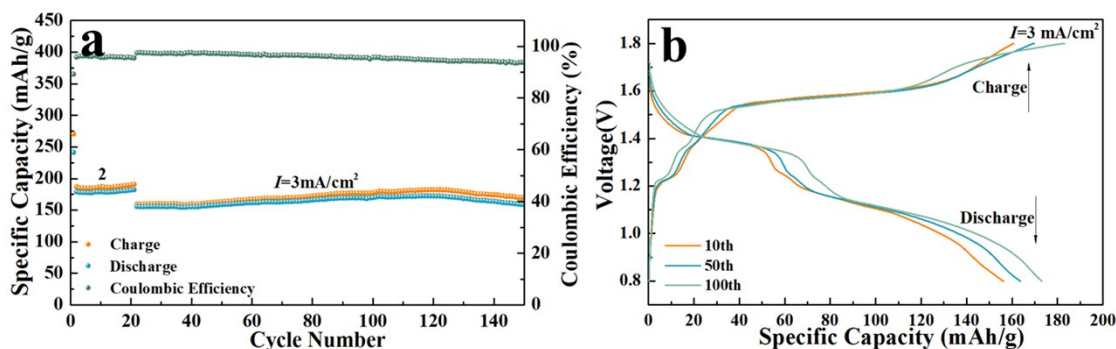
**Figure S6.** (a) Cycling performance of the Fe-FeCl<sub>2</sub>-CR-60 electrode with enhanced mass loading in Fe/FeCl<sub>2</sub>-graphite molten salt battery. (b) The corresponding voltage curves of the Fe-FeCl<sub>2</sub>-CR-60 electrode with enhanced mass loading.



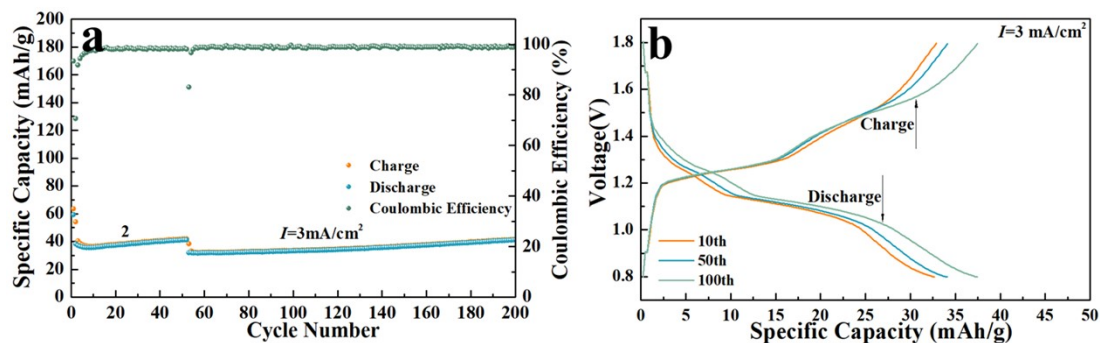
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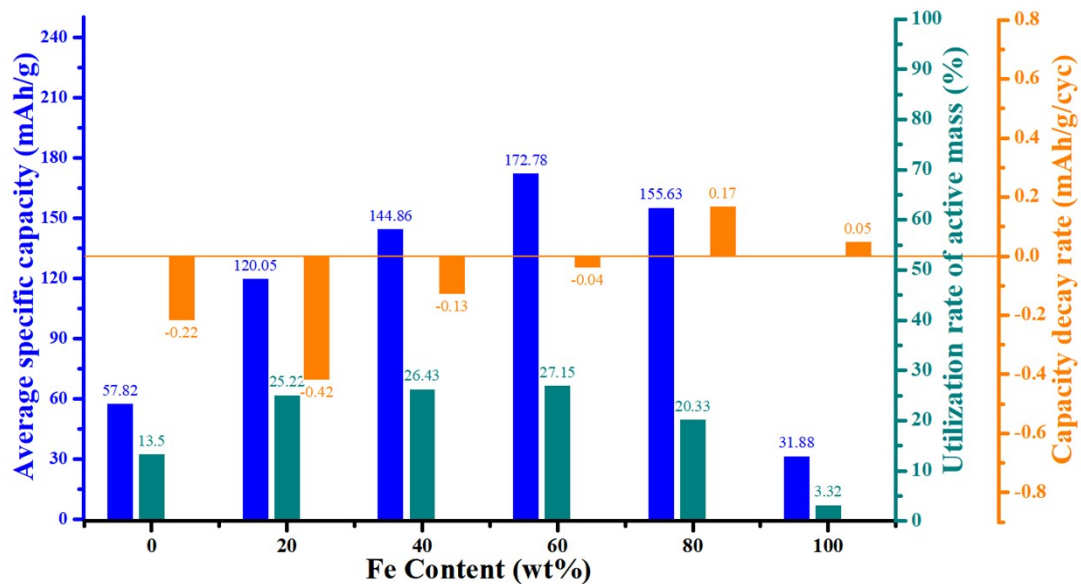
**Figure S8.** (a) Cycling performance of the Fe-FeCl<sub>2</sub>-CR-40 electrode in Fe/FeCl<sub>2</sub>-Graphite molten salt battery. (b) The corresponding voltage curves of the Fe-FeCl<sub>2</sub>-CR-40 electrode.



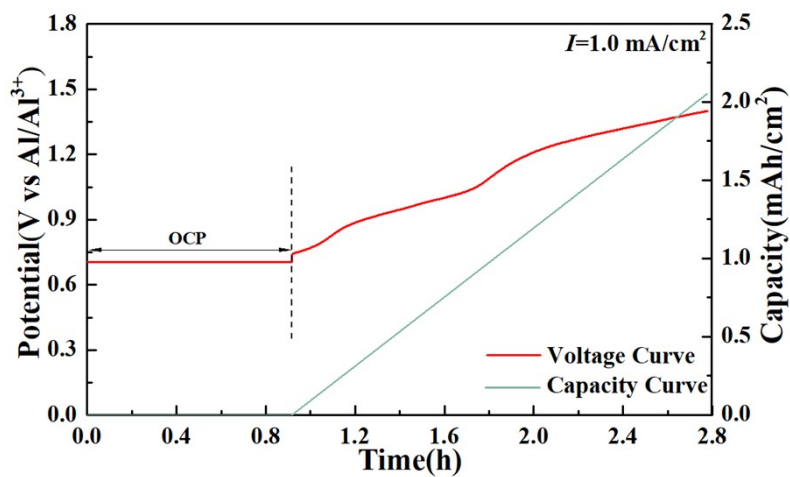
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**Figure S10.** (a) Cycling performance of the Fe-CR electrode in Fe/FeCl<sub>2</sub>-Graphite molten salt battery. (b) The corresponding voltage curves of the Fe-CR electrode.



**Figure S11.** The electrochemical characteristics of Fe-FeCl<sub>2</sub>-CR electrodes with different Fe contents.



**Figure S12.** Galvanostatic chlorination curves of Fe-CR electrode.

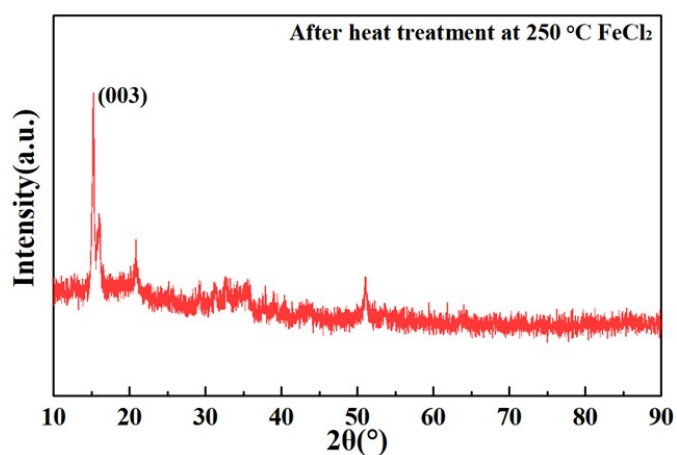


Figure S13. XRD pattern of  $\text{FeCl}_2$  material.

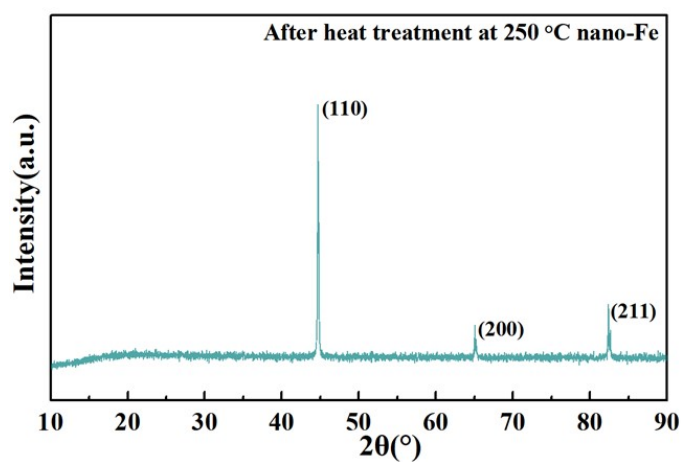


Figure S14. XRD pattern of nano-Fe powder material.

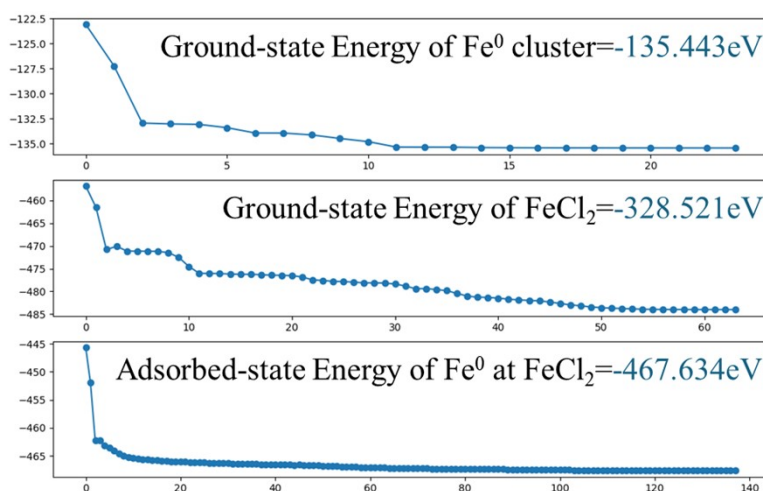
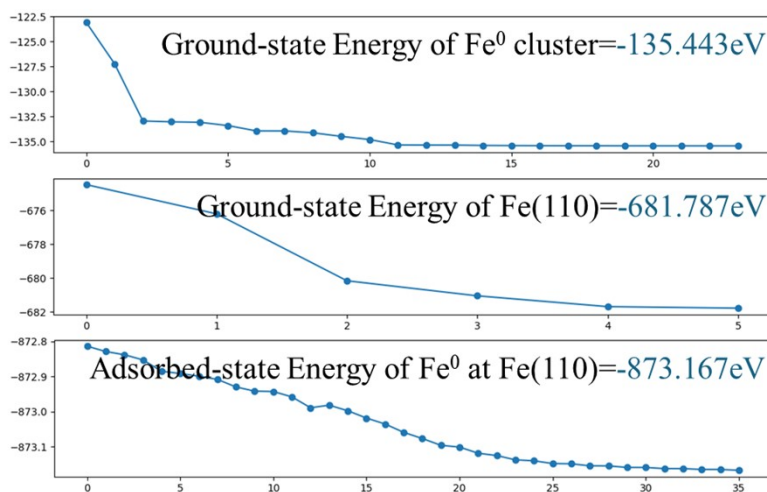
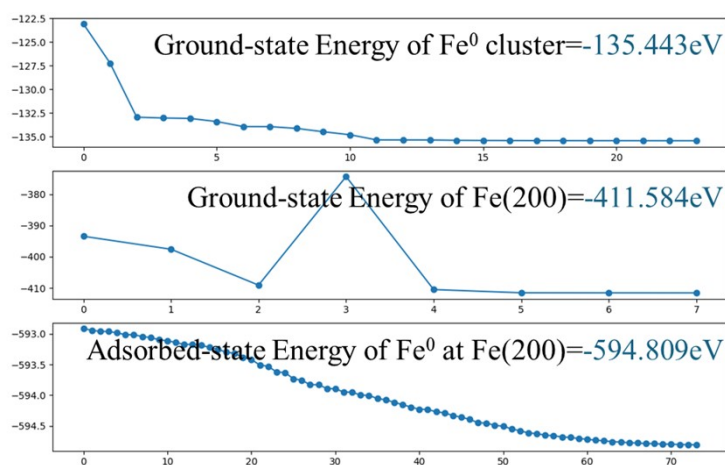


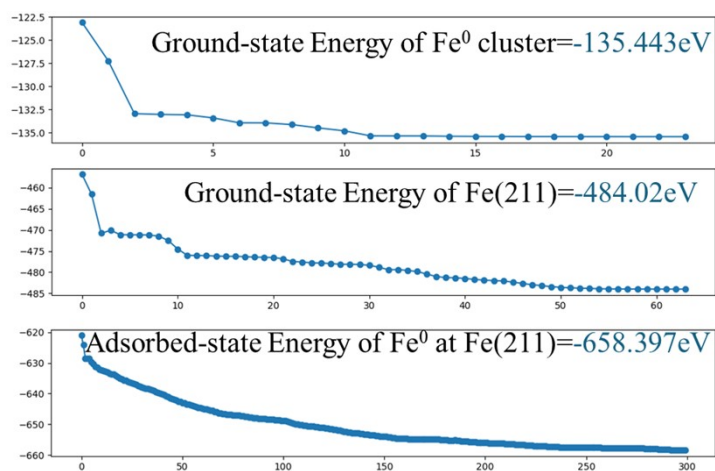
Figure S15. The original chart of the adsorbed energy calculation ( $-3.67\text{eV}$ ) of  $\text{Fe}^0$  cluster at  $\text{FeCl}_2$  (003).



**Figure S16.** The original chart of the adsorbed energy calculation (-55.937eV) of Fe<sup>0</sup> cluster at Fe (110).



**Figure S17.** The original chart of the adsorbed energy calculation (-47.782eV) of Fe<sup>0</sup> cluster at Fe (200).



**Figure S18.** The original chart of the adsorbed energy calculation (-38.934 eV) of Fe<sup>0</sup> cluster at Fe(211).