

# Mechanism of Improving Cycle Capability of $\text{Li}_2\text{S}$ -FeS Composite Positive Electrode Materials by Surface Coating

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High energy density batteries have been required for the next generation EV applications, triggering the search for sulfur-based positive electrode materials with high capacity. Recently, we have developed the Fe-containing  $\text{Li}_2\text{S}$ -based materials ( $\text{Li}_8\text{FeS}_5$ ), and found that  $\text{Li}_8\text{FeS}_5$  cell showed relatively high initial discharge capacity ( $> 700 \text{ mAh}\cdot\text{g}^{-1}$ ) [1]. However, a rapid capacity degradation was observed during the initial several cycles, probably because of some reactions between the electrolyte and the  $\text{Li}_8\text{FeS}_5$ . In the present work, we tried to improve its cycle performances by coating the surface of  $\text{Li}_8\text{FeS}_5$  to suppress the interface reaction.

The  $\text{Li}_8\text{FeS}_5$  was prepared based on the previously reported method [1]. Because of the sensitivity of  $\text{Li}_2\text{S}$  to moisture, all the material synthesis procedures were carried out in an argon atmosphere. The obtained  $\text{Li}_8\text{FeS}_5$  and commercially available titanium tetrachloride ( $\text{TiCl}_4$ ) were mixed in solvent and filtered after stirring, and washed by dimethyl carbonate, according to the previously reported procedure [2]. Then this intermediate product was heated at  $400^\circ\text{C}$  to obtain the sample (hereafter noted as  $\text{Li}_8\text{FeS}_5\text{-Ti}$ ). The electrochemical charge/discharge tests were carried out with  $1 \text{ mol dm}^{-3}$  lithium hexafluorophosphate in 1:9 (v/v) ethylene carbonate and propylene carbonate electrolyte at a current density of  $98.6 \text{ mA g}^{-1}$  (corresponding to 0.1 C) and between 2.6 and 1.0 V vs.  $\text{Li/Li}^+$ .

The  $\text{Li}_8\text{FeS}_5\text{-Ti}$  cells exhibited improved cycle performance in Fig.1. The higher plateau at the discharge could be related to the sulfur redox and the lower plateau are likely related to the iron redox [1]. The capacity corresponding to lower plateau (Q2) decreased with cycling for both sample cells. In contrast, the degradation of the capacity corresponding to the higher plateau (Q1) were suppressed significantly after the surface coating. Therefore we deduced that the surface reaction of  $\text{Li}_8\text{FeS}_5$  with the electrolyte corresponding to the sulfur redox, which could be the main reason for the cycle degradation, may have been suppressed by coating with Ti-containing layer. We are currently examining the in-depth mechanisms of the improved cycle capability by such Ti-containing layer, and the results will be presented in this conference.

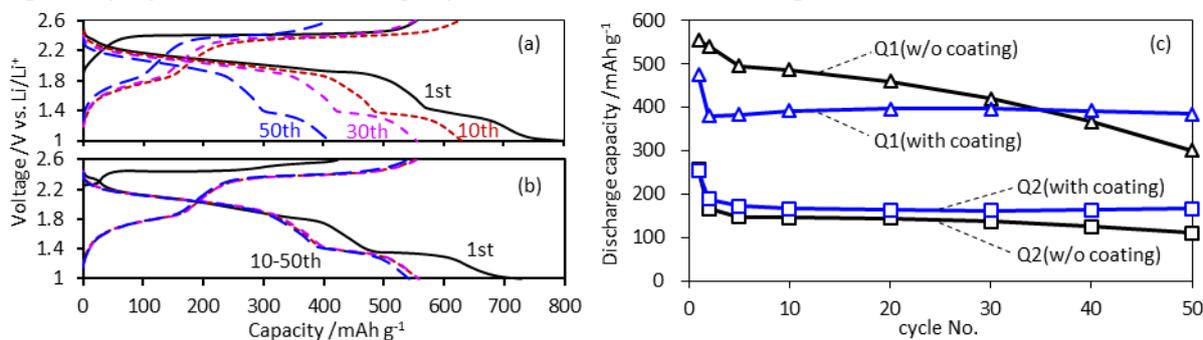


Fig.1 Charge-discharge curves of (a)  $\text{Li/Li}_8\text{FeS}_5$ - (without coating) cell and (b)  $\text{Li/Li}_8\text{FeS}_5\text{-Ti}$  coating cell. (c) Cycling performance of Q1 and Q2 without coating (black) and with coating (blue).

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## References:

1. T. Takeuchi, et.al., J. Electrochem. Soc. 162(9), A1745 (2015).
2. Z. W. Seh, et.al., Nature Communication, 5, 5017 (2014), DOI:10. 1038/ncomms6017