

Supporting information

Dual Lithium Storage of Pt electrode: Alloying and Reversible Surface Layer

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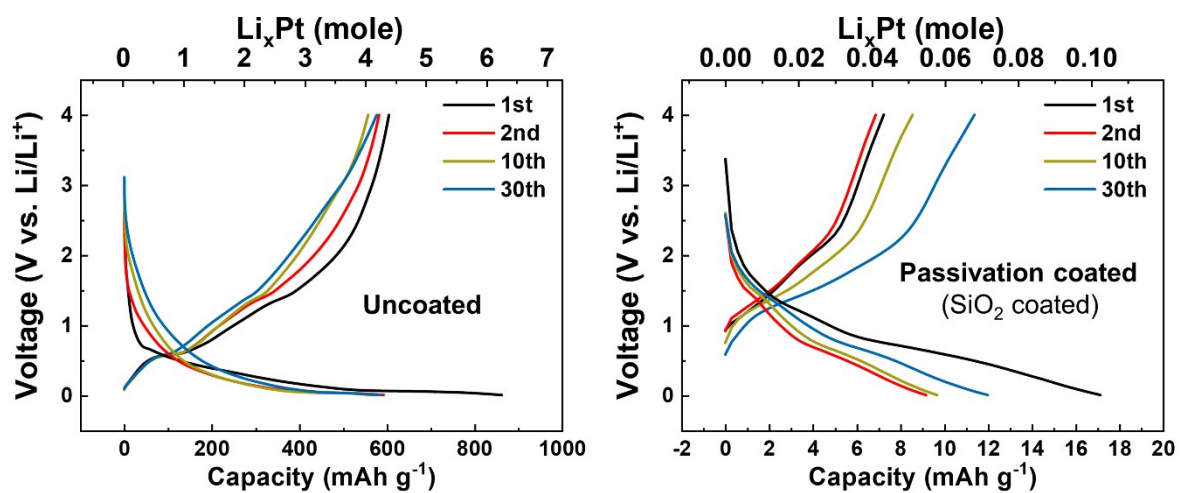


Figure S1. Voltage profile of the uncoated Pt electrode (left), and passivation coated Pt electrode (right) with the potential window from 0.001 to 4.0 V vs. Li/Li⁺ upon the 30 cycles.

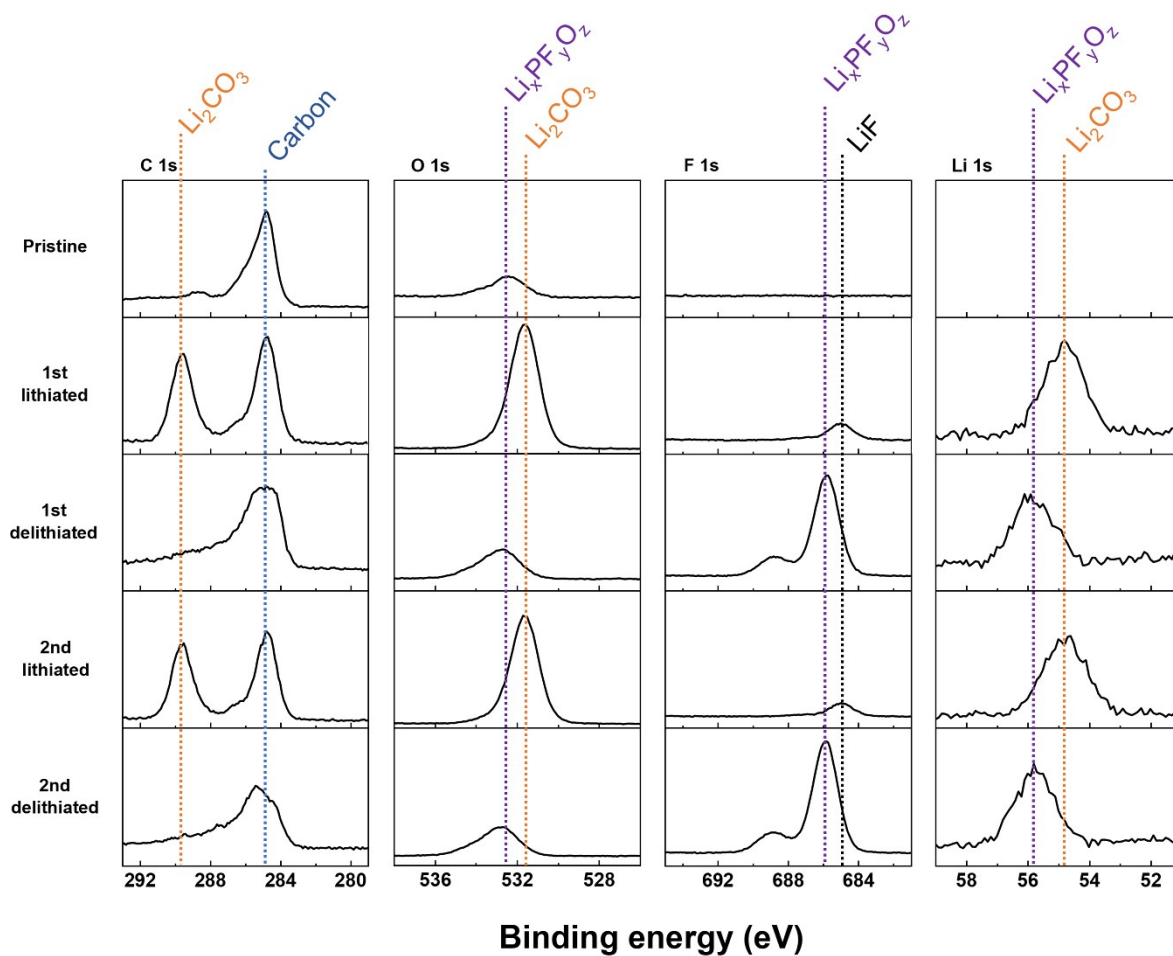


Figure S2. The *ex situ* X-ray photoelectron spectroscopy obtained from the Pt electrode during the 2nd cycles.

Table S1. Theoretical potential of the Pt electrode during the lithiation (Pt → Li₂Pt), and delithiation (Li₂Pt → LiPt → Li_{0.5}Pt → Pt).

	First lithiation	
	Theoretical potential	Experimental value
Pt → Li₂Pt	0.8185 V	0.1055 V (0.7862 V after 2cyc)
	First delithiation	
	Theoretical potential	Experimental value
Li₂Pt → LiPt	0.4167 V	0.5559 V
LiPt → Li_{0.5}Pt	1.1445 V	1.0594 V
Li_{0.5}Pt → Pt	1.2961 V	1.4111 V

The final energy values of each compound of Pt, Li_{0.5}Pt, LiPt, Li₂Pt, and bcc Li obtained through the generalized gradient approximation (GGA) method were utilized.¹ And referring to the following formula proposed by Urban et al.,² the theoretical potential of stepwise reaction constituting the lithiation/delithiation process of the Pt electrode are calculated and compared with the experimental values.

$$V = - \frac{E(Li_{x_2}Pt) - E(Li_{x_1}Pt) - (x_2 - x_1)E(Li^{bcc})}{(x_2 - x_1)F}$$

V : Theoretical redox potential of specific reaction.

$E(Li_{x_2}Pt)$, $E(Li_{x_1}Pt)$, $E(Li^{bcc})$: Internal energies of the lithiated, delithiated Pt phase, and metallic Li.

x_2 : Amount of Li at the lithiated state.

x_1 : Amount of Li at the delithiated state.

F : Faraday constant.

Reference

- 1 A. Jain, G. Hautier, S. P. Ong, C. J. Moore, C. C. Fischer, K. A. Persson and G. Ceder, *Phys. Rev. B*, 2011, **84**, 045115.
- 2 A. Urban, D.-H. Seo and G. Ceder, *npj Comput. Mater.*, 2016, **2**, 16002.