

Structural Change of Li-Graphite Intercalation Compounds

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Introduction Graphite has been used as a negative electrode material in Li-ion batteries. For future development of Li-ion batteries, it is important to identify how Li is intercalated into or de-intercalated from graphite under practical operation [1]. Also, it is interesting to know whether theoretical calculations correctly reproduce experimental facts and, moreover, provide information which is difficult to be predicted by experiments. Although phase stability and voltage profile for Li_xC_6 are already discussed in a theoretical manner [2-3], we reinvestigated them with more systematic way. As a consequence, we obtained different results from them.

Results Firstly, a set of structures which are likely to be stable were selected by classifying in-plane Li ordering (**Fig. 1**) and, then, changing a number of graphene layers between the Li layers. Secondly, we calculated the formation energies of each Li_xC_6 defined as

$$E_{\text{form}} = E(\text{Li}_x\text{C}_6) - xE(\text{LiC}_6) - (1 - x)E(\text{C}_6); \quad 0 \leq x \leq 1$$

It is possible to know from the E_{form} that not only the stable structures appearing upon Li intercalation but how stage number changes or at what x graphene layers are transformed from AB to AA stacking arrangements. Finally, a voltage profile of Li-ion batteries was computed (**Fig. 2**) as

$$\bar{V} = -\frac{E(\text{Li}_{x_2}\text{C}_6) - E(\text{Li}_{x_1}\text{C}_6) - (x_2 - x_1)E(\text{Li})}{x_2 - x_1}; \quad x_2 > x_1$$

which shows good agreement with experiments [4].

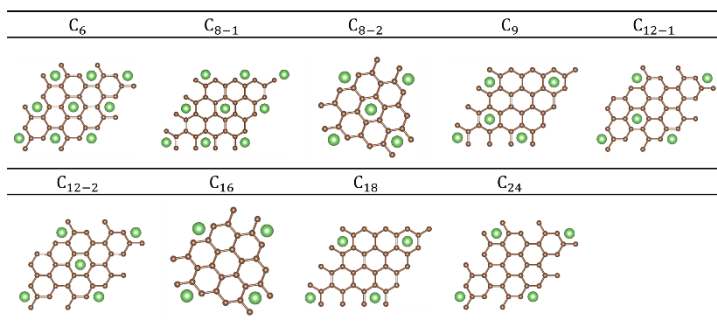


Fig. 1 Classification of in-plane Li ordering.

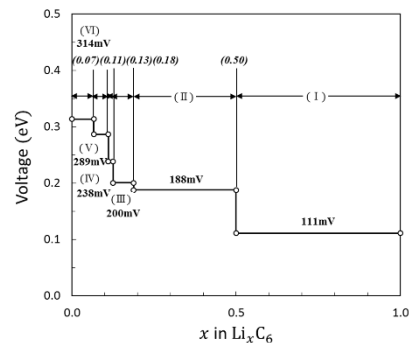


Fig. 2 Voltage profile of Li-ion batteries.

- [1] S. Taminato *et al.*, *Sci. Rep.*, **6**, 28843 (2016). [2] K. Persson *et al.*, *Phys. Rev. B.*, **82**, 125416 (2010). [3] E. Hazrati *et al.*, *Phys. Rev. B.*, **90**, 155448 (2014). [4] T. Ohzuku *et al.*, *J. Electrochem. Soc.*, **140**, 2490 (1993).