

### Non-olivine LiFePO<sub>4</sub> with alluaudite structure for Li rechargeable battery

Jongsoon Kim<sup>1</sup>, Hyungsub Kim<sup>1</sup>, Inchul Park<sup>1</sup>, Young-Uk Park<sup>1</sup>, Jung-Keun Yoo<sup>2</sup>, Kyu-Young Park<sup>1</sup>, Seongsu Lee<sup>2</sup>, and Kisuk Kang<sup>1\*</sup>

<sup>1</sup>Department of Materials Science and Engineering, Seoul National University, 599 Gwanak-ro, Gwanak-gu, Seoul 151-742, Republic of Korea

<sup>2</sup>Department of Materials Science and Engineering, KAIST, 373-1 Guseong-dong, Yuseong-gu, Daejeon 305-701, Republic of Korea

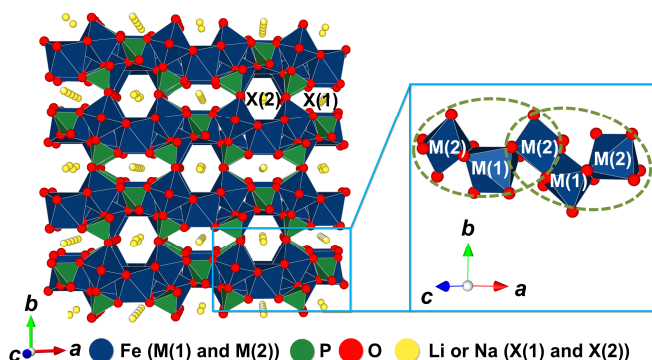
<sup>3</sup>Korea Atomic Energy Research Institute, P.O. Box 105, Yuseong-gu, Daejeon 305-600, Republic of Korea

The combination of naturally abundant Fe and the stable covalent polyanion unit (PO<sub>4</sub>)<sup>3-</sup> in LiFePO<sub>4</sub> is an important factor in the production of cost-effective, safe electrode materials for next-generation lithium ion batteries.<sup>1-4</sup> Since the unprecedentedly high power capability was reported for a LiFePO<sub>4</sub> electrode in 2002,<sup>2</sup> LiFePO<sub>4</sub> has continued to surprise researchers with its unusual physico-chemical characteristics.<sup>5,6</sup> Herein, we further explored the LiFePO<sub>4</sub> chemistry by introducing a novel non-olivine LiFePO<sub>4</sub> with an alluaudite crystal structure. We found that the alluaudite LiFePO<sub>4</sub> shows promising electrochemical properties allowing reversible extraction and insertion of *ca.* 0.8 Li<sup>+</sup> in the structure with one-phase based reaction.

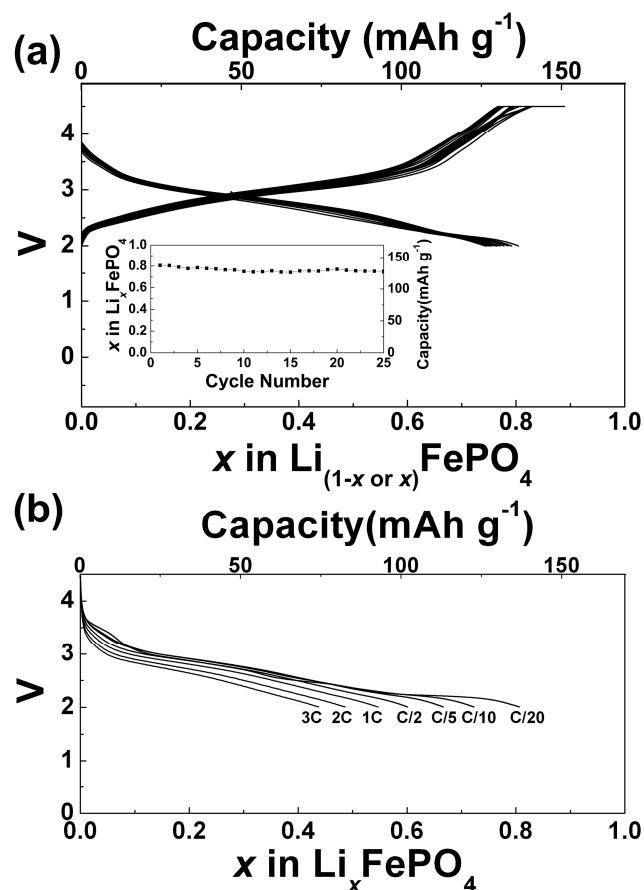
A novel non-olivine LiFePO<sub>4</sub> with an alluaudite structure was successfully prepared, and its structure was investigated in detail through combined ND and XRD analyses. The alluaudite LiFePO<sub>4</sub> showed fundamentally different electrochemical behavior from that of the well-known olivine LiFePO<sub>4</sub>, while the same Fe<sup>2+</sup>/Fe<sup>3+</sup> redox reaction occurred. Almost all of the lithium ions in LiFePO<sub>4</sub> could be reversibly extracted and inserted at a reasonably fast rate, qualifying it as another possible iron-based cathode for lithium rechargeable batteries. We believe that this study will provide insight into the relationship between the structure and the electrochemical activity of LiFePO<sub>4</sub> chemistry.

#### Reference

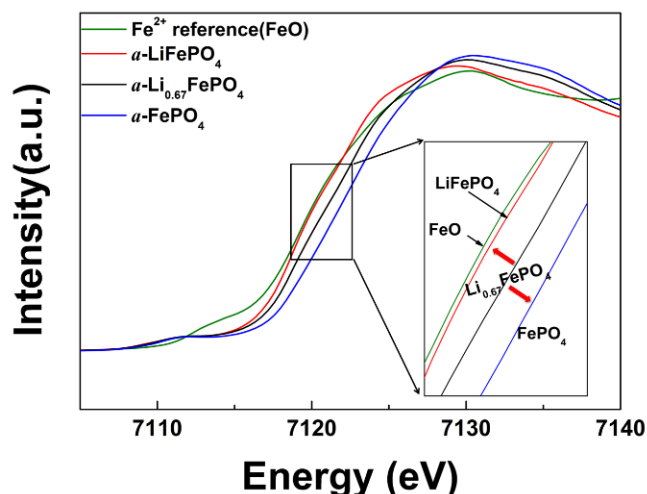
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**Figure 1.** Schematic representation of alluaudite LiFePO<sub>4</sub>. The local geometry of the chains of edge-shared FeO<sub>6</sub> octahedra linked by tetrahedral PO<sub>4</sub> units in the alluaudite.



**Figure 2** (a) Galvanostatic curves of alluaudite LiFePO<sub>4</sub> during 25 cycles at C/20 (inset: cyclability of alluaudite LiFePO<sub>4</sub> during 25 cycles at C/20), (b) discharge curves of alluaudite LiFePO<sub>4</sub> as a function of C rate from C/20 to 3C.



**Figure 3** Normalized Fe K-edge XANES spectra for alluaudite Li<sub>x</sub>FePO<sub>4</sub>.