



# Facial-shape controlled precursors for lithium cobalt oxides and the electrochemical performances in lithium ion battery



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## HIGHLIGHTS

- Two types of LiCoO<sub>2</sub> were synthesized from two cobalt sources (Co<sub>3</sub>O<sub>4</sub>) with different facial-shapes using solid state synthesis.
- Synthesis mechanism of LiCoO<sub>2</sub> was investigated by various experimental techniques.
- Electrochemical performances of LiCoO<sub>2</sub> are strongly dependent on its internal structure.
- Better performances can be achieved just by controlling the facial-shape of precursors for the cathode materials.

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## ABSTRACT

Two types of lithium cobalt oxides (LiCoO<sub>2</sub>) as cathode materials for lithium ion batteries are synthesized from two cobalt sources of different facial-shapes (octahedral and truncated-octahedral Co<sub>3</sub>O<sub>4</sub>) and Li<sub>2</sub>CO<sub>3</sub> using solid state synthesis. From X-ray diffraction and scanning electron microscopy measurements, the reaction mechanism of the formation of LiCoO<sub>2</sub> is investigated. It is revealed that LiCoO<sub>2</sub> from octahedral Co<sub>3</sub>O<sub>4</sub> with only {111} surfaces grows in one direction whereas the crystal orientation of LiCoO<sub>2</sub> from truncated-octahedral Co<sub>3</sub>O<sub>4</sub> with {111} and {100} surfaces is not unique and the spinel intermediates of Li<sub>x</sub>Co<sub>2</sub>O<sub>4</sub> are formed during synthesis. They show largely unequal rate and cycling performances for lithium ion battery, even though their outer appearances are nearly identical. Almost single-crystalline LiCoO<sub>2</sub> from octahedral precursors shows much better electrochemical performances than LiCoO<sub>2</sub> from truncated-octahedral precursors as a lithium ion battery cathode. By studying crystal orientation, it is shown that the poor electrochemical performances of LiCoO<sub>2</sub> from truncated-octahedral Co<sub>3</sub>O<sub>4</sub> are originated by crystal-mismatch between crystallites.

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## 1. Introduction

Lithium cobalt oxide (LiCoO<sub>2</sub>) has been one of the most predominant cathode materials for lithium ion batteries (LIBs) since the first successful commercialization by SONY [1]. For better electrochemical performances, a lot of refinements, such as, dopings [2–5], coatings [5–13], and structural modifications [14–16] in LiCoO<sub>2</sub>, have been attempted. In addition, a few studies have reported that the exterior morphology or shape of LiCoO<sub>2</sub> particles take effect on the electrochemical properties. For example, Li et al. controlled the shape of LiCoO<sub>2</sub> nanocrystals from morphology-tuned CoO precursors [17]. They showed that two different-shaped nanocrystals, i.e. polyhedral and spherical LiCoO<sub>2</sub>

particles, exhibited different electrochemical performances, indicating that lithium intercalation/deintercalation dynamics might be crystal face-sensitive. The same research group also synthesized LiCoO<sub>2</sub> nanowires, which had the one-dimensional nanostructure and exposure of (010) planes, from Co<sub>3</sub>O<sub>4</sub> nanowires as precursors [18]. They showed that these LiCoO<sub>2</sub> nanowires exhibited the good electrochemical performances, especially, the rate capability. In addition, Wei et al. recently showed that a flake-like LiCoO<sub>2</sub> cathode, which had a preferential crystallographic orientation for Li<sup>+</sup> ion migration, demonstrated good reversibility at moderate charging/discharging rates [19]. Besides LiCoO<sub>2</sub>, the outer morphology or shape effects of several other materials for anode as well as cathode on the electrochemical performances have been investigated [20–22].

Meanwhile, there have been a few studies reporting that the crystallinity of the electrode materials has a large influence on electrochemical performances. Nakajima et al. developed new

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synthetic routes for  $\text{LiCoO}_2$  mesocrystals with porous and single-crystalline structures, which exhibited enhanced charge/discharge cycle stability and rate performance [23]. Similarly, Zhang et al. reported that lowering crystal-mismatch, that is, securing high single-crystallinity, is very critical for developing promising electrode materials [24].

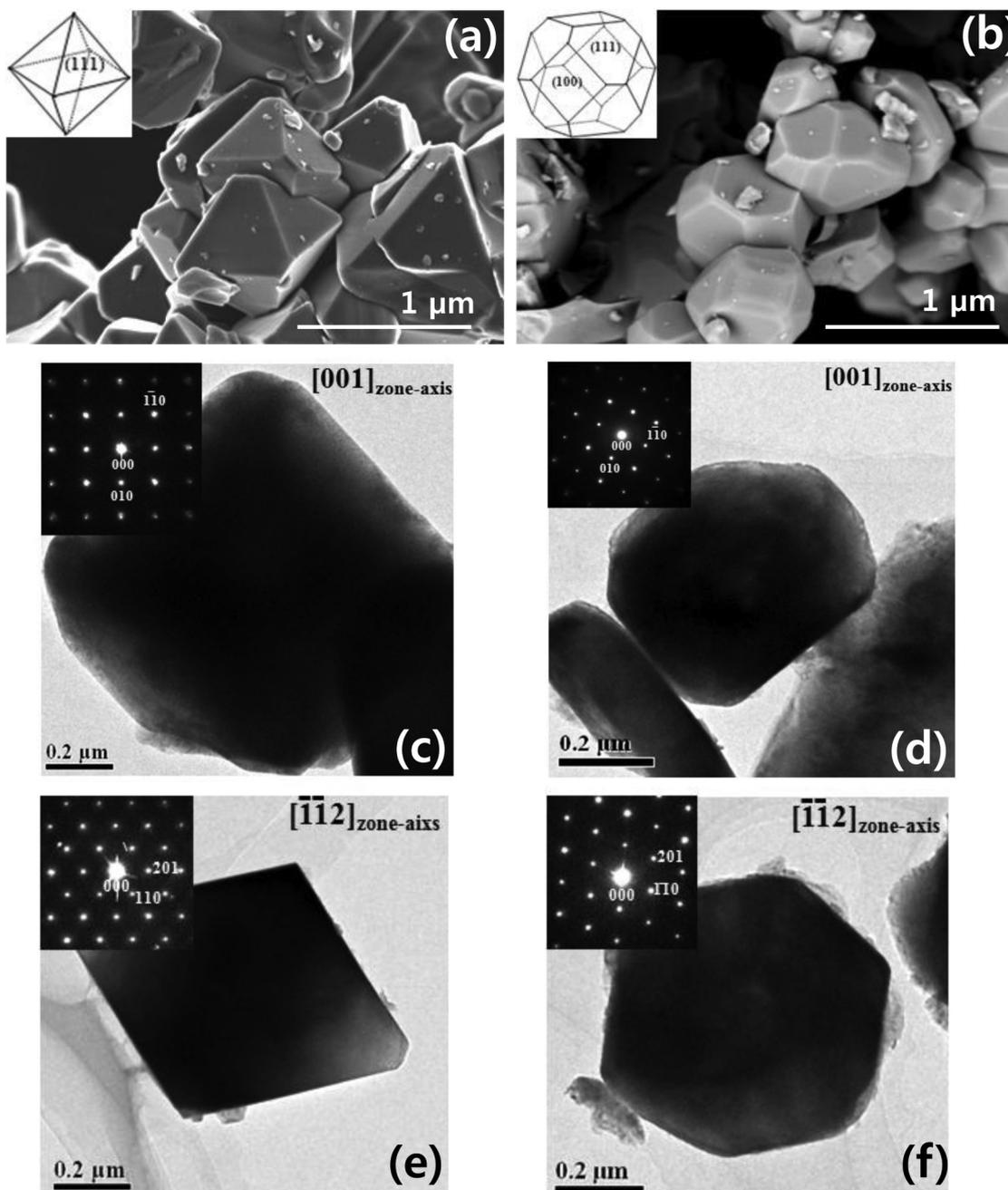
In this study, we described synthesis of two types of  $\text{LiCoO}_2$  crystals, which have different internal fine-structures, from two different  $\text{Co}_3\text{O}_4$  precursors as a cobalt source with different facial-shapes (octahedral and truncated-octahedral). The  $\text{LiCoO}_2$  crystals were elaborately characterized using various experimental techniques in the perspective of structure and electrochemistry. Moreover, this work highlights the relationship between the

electrochemical performances of a cathode material for LIB and its precursors.

## 2. Experimental section

### 2.1. Preparation of $\text{Co}_3\text{O}_4$ precursors

We followed Xiao et al.'s procedure [22]. The facial shape of  $\text{Co}_3\text{O}_4$  is only dependent on the concentrations of reactants. In common,  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  and  $\text{NaOH}$  were dissolved in distilled water. The mixed reactants were heated at  $180^\circ\text{C}$  for 5 h and then cooled to room temperature. After washing with ethanol and water several times, the obtained products were dried at  $60^\circ\text{C}$  for 12 h.



**Fig. 1.** SEM images of (a) octahedral and (b) truncated-octahedral  $\text{Co}_3\text{O}_4$ . 2D TEM images along  $[001]$  zone axis of (c) octahedral and (d) truncated-octahedral  $\text{Co}_3\text{O}_4$ , and along  $[\bar{1}\bar{1}2]$  zone axis of (e) octahedral and (f) truncated-octahedral  $\text{Co}_3\text{O}_4$ .

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