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Carbon-free Mn-doped LiFePO_4 cathode for highly transparent thin-film batteries

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HIGHLIGHTS

- Exploration of carbon-free transparent $\text{LiFe}_{1-x}\text{Mn}_x\text{PO}_4$ thin films by CCS.
- The capacity of carbon-free $\text{LiFe}_{0.77}\text{Mn}_{0.23}\text{PO}_4$ thin films is $45.7 \mu\text{A h/cm}^2 \cdot \mu\text{m}$.
- Transmittance of Carbon-free $\text{LiFe}_{0.77}\text{Mn}_{0.23}\text{PO}_4$ thin film exhibits 82%.
- Carbon-free $\text{LiFe}_{0.77}\text{Mn}_{0.23}\text{PO}_4$ is suitable for transparent thin film batteries.

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ABSTRACT

The search for transparent battery cathodes primarily focuses on patterned electrodes with feature sizes below the optical absorption limit. This significantly limits the electrode capacity, as a large electrode area remains unused to maintain transparency. Herein, we report transparent olivine $\text{LiFe}_{0.77}\text{Mn}_{0.23}\text{PO}_4$ thin-film electrodes discovered through high-throughput continuous-composition-spread sputtering. After investigating six different Mn doping ratios, we found the optimal Mn-doped olivine composition with an enhanced discharge capacity of $45.7 \mu\text{A h/cm}^2 \cdot \mu\text{m}$ without using excessive nanosized features or carbon coating. The thin-film electrode exhibits a clear redox activity for both $\text{Fe}^{3+/2+}$ and $\text{Mn}^{3+/2+}$, resulting in an enhanced average voltage over LiFePO_4 composition. A 250-nm-thick film exhibits an optical transmittance of over 80% in the visible region. The results in this study demonstrates that transparent cathode thin films can be developed based on phospho-olivines via doping strategies with high-throughput continuous-composition-spread sputtering methods.

1. Introduction

Transparency enables unique applications in electronics. Recent research effort for transparent electronics has resulted in interest in transparent batteries, often considered the most difficult element to achieve transparency [1]. The successful fabrication of durable and transparent batteries may enable a new generation of portable electronics in which the entire device is transparent. Applications such as electronic contact lenses may be powered by an onboard battery while

ensuring the lens' transparency [2–5].

The development of transparent batteries often relies on patterning electrodes to feature sizes below optical absorption lengths [1]. Patterning nontransparent materials, however, results in reduced energy density, as a large portion of the electrode area becomes unutilized to achieve transparency. For patterned electrodes, 25% of active area per unit area can be used [1]. Similar strategies to fabricate transparent thin-film batteries have been reported by Oukassi et al. with non-transparent materials [6]. Oukassi et al. reported that 34% of active area

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