High-Rate Blended Cathode with Mixed Morphology for All-Solid-State Li-ion Batteries

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Response to Reviewer comments

#1 reviewer

This manuscript reports blended (LFP/LVP) cathode formulations for all-solid-state batteries and their improved electrochemical performance such as rate-capability and cycling stability. It is of significance to come up with an approach to realizing high-performance electrodes for solid-state battery chemistries, and the manuscript deals with this important topic. Furthermore, the authors clearly demonstrated improvement in electrochemical performance of the LFP/LVP-blended electrode over the conventional one. I recommend publishing this manuscript with minor revisions as commented below:

Q 1. Most of electrochemical performances were evaluated at 70°C. Provide the reason why the measurements were made at a specific temperature of 70°C and discuss the possible experimental findings at room temperature.

A 1. PEO binder has low ionic conductivity because of crystalline phase presenting at below melting points of PEO (65°C), which hinder the operation of all-solid-state Li-ion batteries (ASLBs) with PEO-based hybrid electrolyte at room temperature. For this reason, electrochemical tests were carried out at 70°C.

2. Explain why the blended LFP/LVP electrode was prepared with a ratio of 8:2. What happens below or beyond that ratio?

A 2. As a reviewer's comment, we have studied on the electrochemical performance according to various (LFP/LVP) weigh ratio such as 10:0, 8:2, 5:5 and 0:10 for finding optimized mixing condition. Among these ratios, in case of 8:2 (LFP/LVP) weigh ratio, it displayed the best electrochemical performance. So, we are investigated the relation between electrochemical performance and blending ratio of LFP:LVP for systematic analysis.

3. This paper demonstrated the improvement of electrochemical properties by blending LVP with LFP. What would be the main reason for the observed improvement --- particle shape or carbon source or both?

A 3. We think that the reason of improved electrochemical performance of blended cathode material is synergy effect for particle shape of the LVP material and carbon coated on cathode surface. One of the problems with LFP cathode material has low electrical conductivity. Contained carbon source LVP cathode material contributes to the enhanced electrical conductivity, and the presence of rod-shaped particles in addition to nano-sized particles can provide better bonding between particles and contribute to improvement of electrochemical performance and we mentioned the main reason in manuscript.

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#2 reviewer

This manuscript presents the effect of blended cathode materials on the performance of all-solid-state batteries with oxide-based organic/inorganic hybrid electrolytes. This manuscript is well organized and provides a practical guideline for reliable all-solid-state batteries, but there are some unclear points to be addressed.

Q 1. In general, LVP cathode material is widely used as a high voltage range up to 4.5 V. However, blended cathode material mixed in LFP and LVP measure only up to 4.0 V. What is the merit of LVP cathode material in blended cathode material?

A 1. As the reviewer's comment, LVP cathode material are mainly known to be used for high voltage system. However, we choose the LVP cathode material for blending of LFP due to their structural stability at elevated temperature unlike layered structure material owing covalently bonded $(PO_4)^{3-}$ units. In case of all solid state battery using PEO binder for organic/inorganic hybrid membrane, electrochemical evaluation has to be at high temperature above 70°C. Therefore, LVP cathode material having great structural stability can attribute to enhance characteristic of LFP cathode material in all solid state battery.

Q 2. In the experimental part, LLZO, cathode material, super P and binder are mixed in manufacturing positive electrode. What is the role of LLZO in the positive electrode?

A 2. In particular, inorganic nanoparticles, such as LATP, play important roles as ionic conducting fillers in PEO-based composite electrolytes that enhance electrochemical properties and thermal stabilities, and protect the lithium dendrite. Similarly, LLZO in the positive electrode improves the lithium ion conductivity of the electrode and provides a path through which lithium ion can move. And we mentioned the role of LLZO in the positive electrode in manuscript.

Q 3. Fig. 7 shows the high rate characteristics of blending and LFP materials. And Fig. 7 (a) shows that the discharge capacity retention of the two materials does not differ significantly between 0.1 C and 0.2 C. Why does not differ?

A 3. As the reviewer's comment, at the low current density LFP and blended cathode material show similar discharge capacity retention. Since LFP material has strong $(PO_4)^{3-}$ bonding, there is almost no structural change as charging and discharging progresses, improving structural stability. Therefore, it is thought that the difference in discharge capacity retention rate does not show much in low current regions such as 0.1 and 0.2 C. And we mentioned the manuscript.