Supporting Information

Li_{0.35}La_{0.55}TiO_3 Nanofibers Enhanced Poly(vinylidene fluoride)-Based Composite Polymer Electrolytes for All-Solid-State Batteries

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Figure S1. (a, c) SEM images of the as-spun precursor nanofibers and LLTO nanofibers. (b, d) Corresponding diameter distributions.

Fig. S1a-d show the SEM images of the as-spun precursor nanofibers and the ceramic LLTO nanofibers and the corresponding diameter distributions. The electrospun precursor nanofibers have a uniform fibrous structure and a smooth surface (Figure S1a) with an average fiber diameter of 2770 nm (Figure S1b). Uniform LLTO nanofibers with a very smooth surface appearance were formed and the average fiber diameter was 740 nm (Figure S1c and d).
Figure S2. Constructed surface models of LLTO nanofiber. (a) top view of (110) surface, (b) side view of (110) surface, (c) top view of (200) surface, (d) side view of (200) surface, (e) top view of (211) surface, (f) side view of (211) surface.
Figure S3. NMP is adsorbed on LLTO nanofibers. Optimized structures for side view of (110) surface (a), side view of (200) surface (b), side view of (211) surface (c).
Figure S4. Digital photograph of the PVDF-CPEs (15 wt. % LLTO nanowires) film at initial (a) and final stretch state (b). These digital photos proof the enhancement of mechanical properties can be attributed to the introduction of LLTO nanofibers.

![Digital photograph of film](image)

Figure S5. Electrochemical characterization of PVDF-LLTO(15 wt.% CPEs in solid-state batteries. (a) Cycling performance of the battery at 1 C. (b) The first three charge/discharge curves of NCM811/PVDF-CPEs/Li battery at 1 C.

The electrochemical performances of NCM811/PVDF-CPEs (15 wt. % LLTO)/Li are also shown in Figure S5. The electrochemical performance of all-solid-state LIBs using LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2 (NCM811) cathode was tested at 1 C. The solid batteries delivered a high initial discharge capacity of 178 mAh g^{-1} at 1 C, and it could run normally at room temperature with a high Coulombic efficiency (CE) of >96% all the way. The results proved that these PVDF-CPEs can match with various cathodes, which again presents the advances of these PVDF-CPEs for possible commercial application.