## A Highly Conductive Quasi-Solid-State Electrolyte Based on Helical Silica Nanofibers for Lithium Batteries

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Figure S1 Chemical structure of gelator *D*-ValPyBr and HR-TEM image of a helical tubular silica nanofiber.

Silica nanofibers were prepared via a classic sol-gel transcription method, using the self-assembly of gelator *D*-ValPyBr [(R)-1-(11-((3-Methyl-1-(octadecylamino)-1-oxobutan-2-yl)amino)-11-oxoundecyl)-pyridin-1-ium bromide] as the template and TEOS as the precursor. The formation process of helical silica nanofibers was as follows: Firstly, the gelator *D*-ValPyBr self-assembled to form single-strand helix gel fibers. Secondly, TEOS hydrolyzed under the help of ammonia (acting as the catalyst) to form silica oligomers, which adsorbed onto the surface of single-strand helical gel fibers by electrostatic action. At the same time, these single-strand helical gel fibers are further self-assembled into multiple-strand helical nanofiber bundles. Finally, silica oligomers were polymerized on the surface of supramolecular assemblies, and helical tubular silica nanofibers were obtained after removing the organic template by calcination. The obtained nanofibers possess a tubular cavity, which will benefit the soaking of IL.



## Figure S2 WAXRD pattern of HSNFs and HSNGE.

The WAXRD pattern of HSNFs indicates that silicas are previously amorphous, only one broad peak at  $2\theta = 22^{\circ}$  is observed. After soaking Li-IL, two broad peaks centered at  $2\theta = 12.5^{\circ}$  and  $19.5^{\circ}$  imply that the obtained HSNGE contains amorphous silica matrix coexisting with the ionic liquid.



Figure S3 Typical TGA curves of HSNGE and ILE.



Figure S4 Impedance spectra of the SS|HSNGE|SS cell acquired at different temperatures.



Figure S5 Initial charge and discharge curves of a LiFePO<sub>4</sub>|HSNGE|Li cell at different current densities.

The discharge plateau voltage decreases with the increase of the current rate, which

is attributed to the enhancement of battery polarization.



Figure S6 (a) Photograph and (b) FE-SEM image of HSNGE after 100 cycles as tested in a LiFePO4|HSNGE|Li cell.

The cell after cycling was taken apart and the photograph of HSNGE was taken. As seen in Fig. S6a, the solid-state electrolyte can still remain self-standing pellet. Helical nanofibers can be seen in Fig. S5b, they still maintain the initial structure, suggesting excellent structural stability.



**Figure S7** Nyquist plots for the LiFePO<sub>4</sub>|HSNGE|Li cell acquired in the frequency range of 100 KHz to 10 MHz: (a) as prepared and (b) after 50 cycles.