# SupportingInformation:

## Novel inorganic tin phosphate gel:Multifunctional material

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## EXPERIMENTAL SECTION

### Synthesis of nanolayered 15Å $H^+$ SnP, $Sn(HPO_4)_2 \cdot 3H_2O$ phase

Five mL of 1M stannic chloride was added dropwise to 20 mL of $NaH_2PO_4$solution (3M) while stirring in the MH vessel. This was the starting solution for the previously reported procedure by us.[1] The above procedure, however, was slightly modified here by adding 60 mmol of NaCl to the above solution so that the total Na ions is double that of the previous experiments [1]. The above suspension was then adjusted to a pH value of 0.4 using concentrated HCl and then treated at 195°C/2h under microwave-hydrothermal (M-H) conditions using a MARS-5 equipment (CEM corporation) with controlled temperature conditions. This system uses 2.45 GHz frequency and has a maximum power of 1200 W, which can be varied from 0 to 100 % and the system can be operated to a maximum temperature of 240°C. After the M-H treatment, the synthetic samples were washed six times with 1M HC1 and then dried at 60°C prior to X-ray diffraction (XRD) analysis for phase identification. The acid washing was given not only to remove excess salts but also to exchange the interlayer $Na^+$ cations with protons as was reported by us[1] and others[2].

Chemical analysis of the 15Å SnP-$H^+$ phase was done by dissolving in HF followed by neutralization with boric acid. Analyses of Sn and P were done by inductively coupled plasma emission spectrometry using a Perkin-Elmer Optima 5300 ICP. No sodium was present. The 15Å SnP-$H^+$ phase contained three layers of water molecules based on its analyzed chemical composition of 1Sn to 2P molar ratio and thermal gravimetric (TGA) analysis, which showed a 14.4%

weight loss amounting to three layers of water molecules in the SnP-$H^+$ phase. All this information was reported in our previous paper in ChemComm and its supplementary information[1].

## Synthesis of nanolayered 15Å $NH_4^+$ SnP phase [3]

A 5ml of 1M SnCl4 and 2.78g urea were added to 6.25g H3PO4 under magnetic stirring. After stirring for 15min, transparent solution was formed. Then the mixture was treated under conventional hydrothermal condition at 200 °C for 2 days. White precipitate was obtained, which was collected by centrifugation and washed 4 times with deionized water and 2 times with ethanol. Then the product was dried in an oven at 65 °C. This 15Å $NH_4^+$ SnP phase, $Sn(NH_4PO_4)(HPO_4)\bullet 2H_2O$ was used here solely for showing the layer structure by high-resolution TEM because it is more stable in the beam compared to 15Å $H^+$-SnP phase. No other experiments were done with this material in this paper. This phase was reported[3] to contain one $NH_4^+$ and one $H^+$ rather than 2 $H^+$ in the case of $Sn(HPO_4)_2\cdot 3H_2O$.

## Instrumental techniques for analyses

Powder XRD analysis was done using a PANalytical Empryean X-Ray Diffractometer with CuKα radiation. High resolution transmission electron microscopy (TEM) was done on a JEOL JEM-2010F microscope operated at 200kV.

## Proton Conductivity Measurement

The proton conductivities of 15Å SnP-$H^+$ phase and Nafion were measured using the same experimental system and method as described in our previous paper [4]. Nafion was used as a standard for proton conductivity in our lab and hence we measured its proton conductivity at 7 humidity levels[4]. However, we commonly used only 3 or 4 humidity levels for testing unknown samples[4,5].

**Electrode preparation and electrochemical measurement.**

Coin cells (CR2032) were fabricated to evaluate the electrochemical properties of SnP material lithium-ion capacitor cathode while using lithium metal as both the counter electrode and reference electrode. Working electrodes wereprepared by coating a slurry mixture of SnP phase, ketjenblack (KB), and polyvinylidene fluoride (PVDF) with a mass ratio of 6:3:1 (using N-methyl-2-pyrrolidone as solvent) on Al foil (lithium-ion capacitor cathode) current collectors. The electrolyte was 1M $LiPF_6$ in ethylene carbonate/dimethyl carbonate/ethyl methyl carbonate (EC/DMC/EMC, 1:1:1 vol%, 1 % VC additive). Before test, the assembled cell was aged for 2 h. Charge–discharge tests were performed on a LAND CT2001A cell tester (Wuhan Lanhe, China) with the potential range of 0.01-3.0 V vs. $Li^+/Li$. Cyclic voltammetry (CV) tests were performed on an electrochemical workstation (CHI 660E).

## References:

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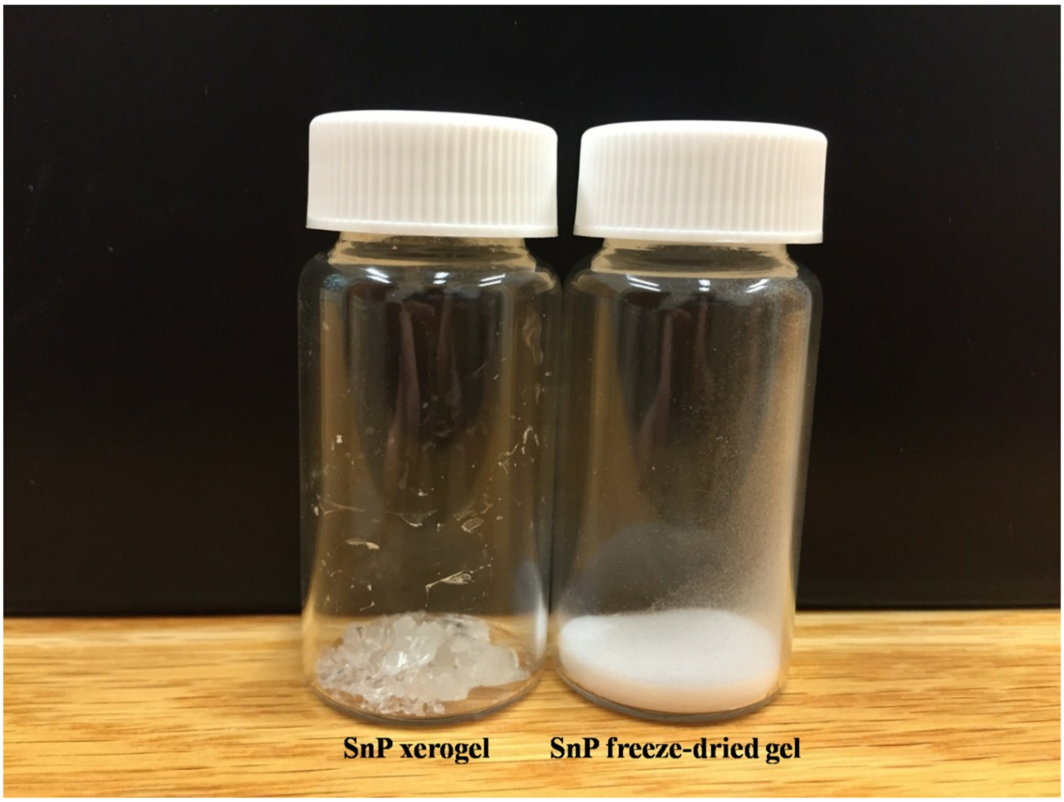

**Fig. S1** SnP transparent gel changed to SnP xerogel upon oven drying and appeared like an opaque silica xerogel while the freeze died transparent gel showed a fluffy white powder of delaminated nanolayers.

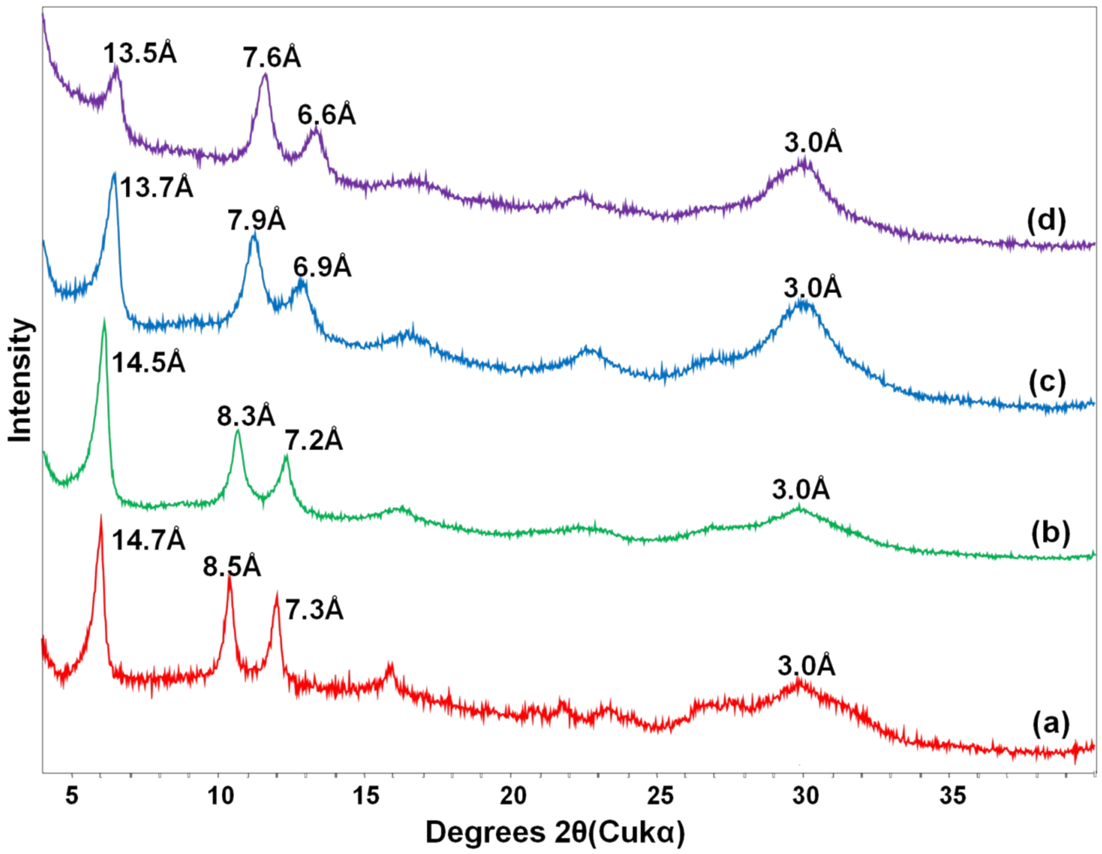

**Fig. S2** Powder XRD patterns of $H^+$-SnP prepared by conventional-hydrothermal process at 220° C/7days and heated at different temperatures to determine dehydration of the interlayers: (a) As-prepared, (b) heated at 200°C for 4hrs, (c) heated at 350°C for 2hrs and (d) heated at 450°C for 2hrs.

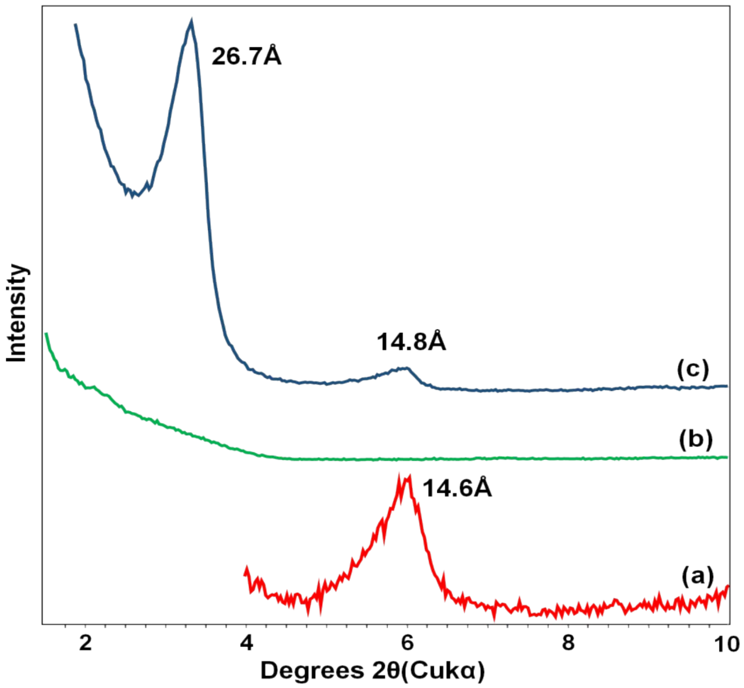

**Fig. S3** As-prepared $H^+$ SnP (a) and the $H^+$ SnP treated with octylamine for several days showing amorphous phase by exfoliation of SnP (b) and showing a 26.7 Ȧ octylamine intercalated SnP phase by self assembly after further exposure to octylamine for several more days (c).

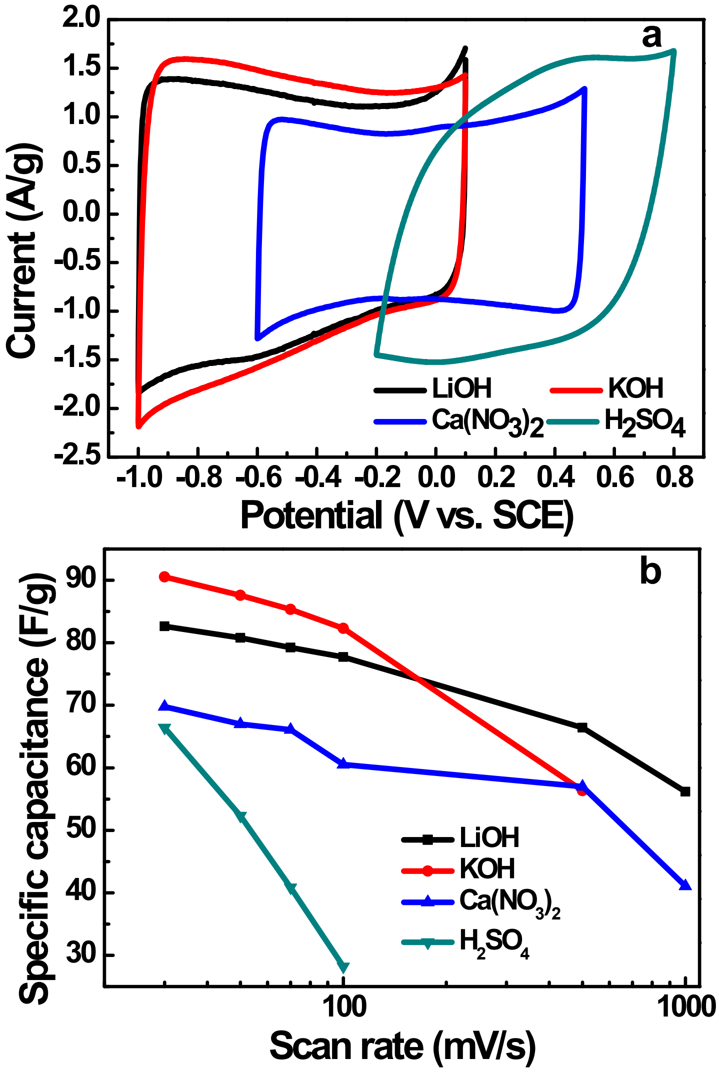

**Fig. S4** Electrochemical performance of SnP freeze-dried gel for supercapacitor electrode in different electrolytes. (a) CV curves at the scan rate of 30 mV/s and (b) Specific capacitance vs scan rate. The electrolytes are 2 M LiOH, 2 M KOH, 1 M $Ca(NO_3)_2$, 0.5 M $H_2SO_4$. The electrochemical tests were performed at room temperature.