Electronic supplementary materials *Mendeleev Commun.*, 2025, **35**, 331–333

Aqueous processed Li-ion battery electrodes with hydrolyzed polyacrylonitrile binder

Artem A. Asharchuk, Aleksei V. Kubarkov, Alexander V. Babkin, Oleg A. Drozhzhin and Vladimir G. Sergeyev

1. Experimental details

Materials

Hydrolyzed polyacrylonitrile (HyPAN, Burkhimsnab, Russia), $M_w = 3 \times 10^5$, contained acrylic acid and acrylamide monomer units in a molar ratio of 75:25[S1] and was purified by reprecipitation. For this purpose, 4 g of HyPAN was dissolved in 200 mL of distilled water; 5 mL of 10 M HCl was added to the prepared solution on a magnetic stirrer. After stirring for 1 h, the precipitate formed was filtered off, washed with distilled water, and dried at 60 °C. The resulting protonated HyPAN was suspended in distilled water (20 mg mL^{-1}), then aqueous solution of LiOH (75 mg mL^{-1}) was added dropwise until HyPAN was dissolved and pH reached 5.

Polyvinylidene fluoride Solef™ 5130 (PVDF, Solvay S.A., Belgium), carboxymethylcellulose (CMC, Gelon LIB, China), aqueous dispersion of styrene-butadiene rubber (SBR, Gelon LIB, China), carbon-coated $LiFePO_4$ (BTR, China), and single-walled carbon nanotubes Tuball™ (SWCNTs, "Tuball", OCSiAl, Russia) were used as received. Double distilled water and *N*-methyl-2-pyrrolidone (Khimmed, Russia) were used as solvents to prepare electrode slurries.

Preparation of the electrodes

Solutions of the polymer binders (20 mg mL^{-1}) were prepared. HyPAN and CMC/SBR (CMC:SBR = 1:1 w/w) were dissolved in water, whereas PVDF was dissolved in NMP. SWCNTs were dispersed in the binder solutions (1.05 mg mL^{-1}) for 10 min by tip-ultrasonication using a Vibra-Cell VCX 750 ultrasonic processor. The resulting dispersions were thoroughly mixed with $LiFePO_4$ powder; the fraction of $LiFePO_4$ was 95 wt.% with respect to the total solids content. The resulting slurries were spread on the surface of carbon-coated aluminum foil current collector (Gelon LIB, China) using a doctor blade applicator (Novotest AU-823) with a 300 μm gap. After being dried in air for 1 day, the coatings were roll-pressed. The disc-shaped electrodes (2 cm^2 area) were cut out of the resulting laminates, weighed with 0.01 mg accuracy, and after that dried under dynamic vacuum at 110 °C for 4 h. The active mass loading of $LiFePO_4$ was *ca.* 4 mg cm^{-2}.

Characterization

To determine the SWCNTs dispersion yield, the SWCNTs-HyPAN dispersions were centrifuged at 12000 rpm for 10 min. After the precipitate was removed, the concentration of SWCNTs in the remaining dispersion was determined by electron spectroscopy using a preliminary determined extinction coefficient of 38.7 mL mg^{-1} cm^{-1} at 500 nm (Figure S4).

Scanning electron microscopy (SEM) images of the electrodes were obtained using a Helios G4 PFIB UXe DualBeam microscope (ThermoFischer Scientific, Walthem, MA, USA)

The electrodes were tested in electrochemical half-cells with lithium metal foil anode, Celgard 2500 microporous polypropylene separator and 1M $LiPF_6$/EC:DMC (1:1 v/v) electrolyte (Sigma Aldrich). The potentiostatic charge/discharge experiments were performed using a P-20X8 potentiostat-galvanostat (Electrochemical Instruments, Russia) within the potential range of 2.0–4.0 V *vs.* Li/Li^+. The capacity values were normalized by the weight of $LiFePO_4$ material. Cyclic voltammetry characterization was carried out at the scan rate of 100 μV s^{-1}.

2. Electrochemical stability of HyPAN binder

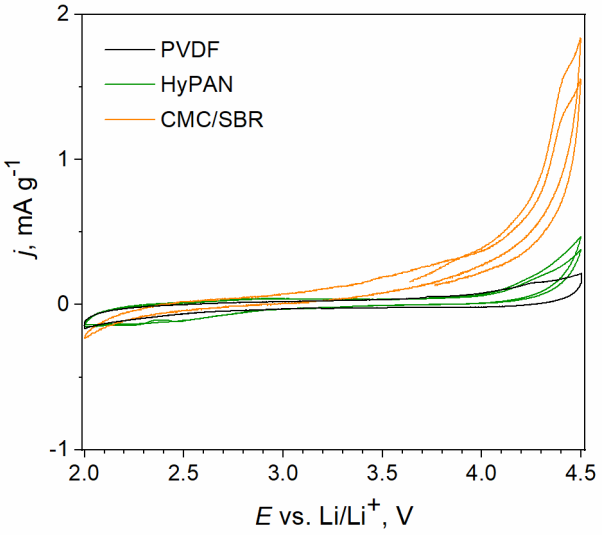

Figure S1 Cyclic voltammograms of the binder films (100 μV s^{-1}).

3. Dispersibility of SWCNTs in HyPAN solutions

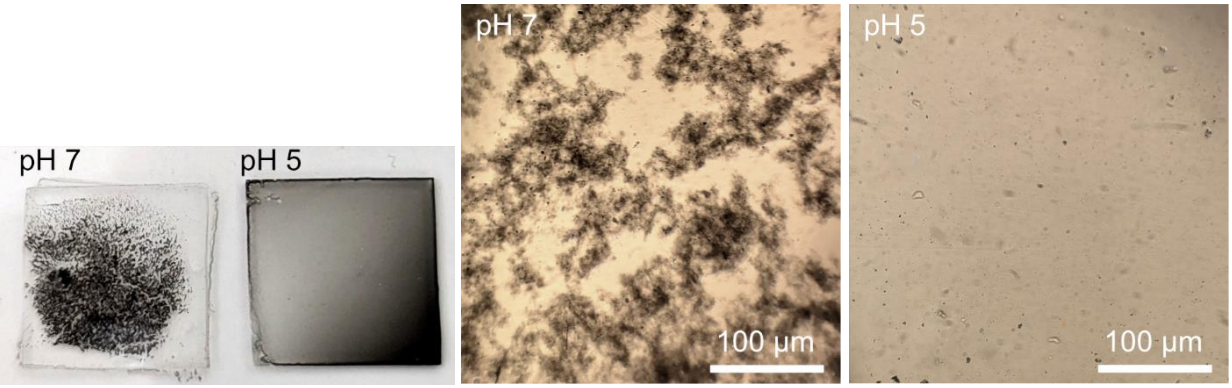

Figure S2 Optical images of the SWCNTs dispersions in HyPAN aqueous solutions at different pH (1.05 mg mL^{-1} of SWCNTs, 20 mg mL^{-1} of HyPAN).

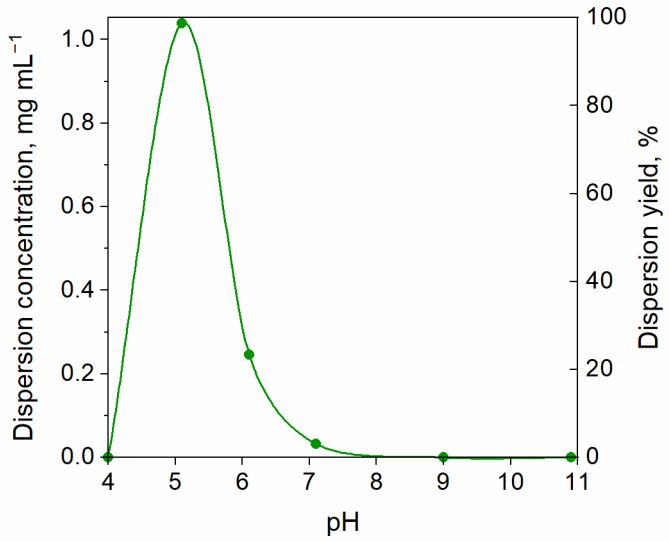

Figure S3 Dispersibility of SWCNT in the aqueous HyPAN solutions as a function of pH.

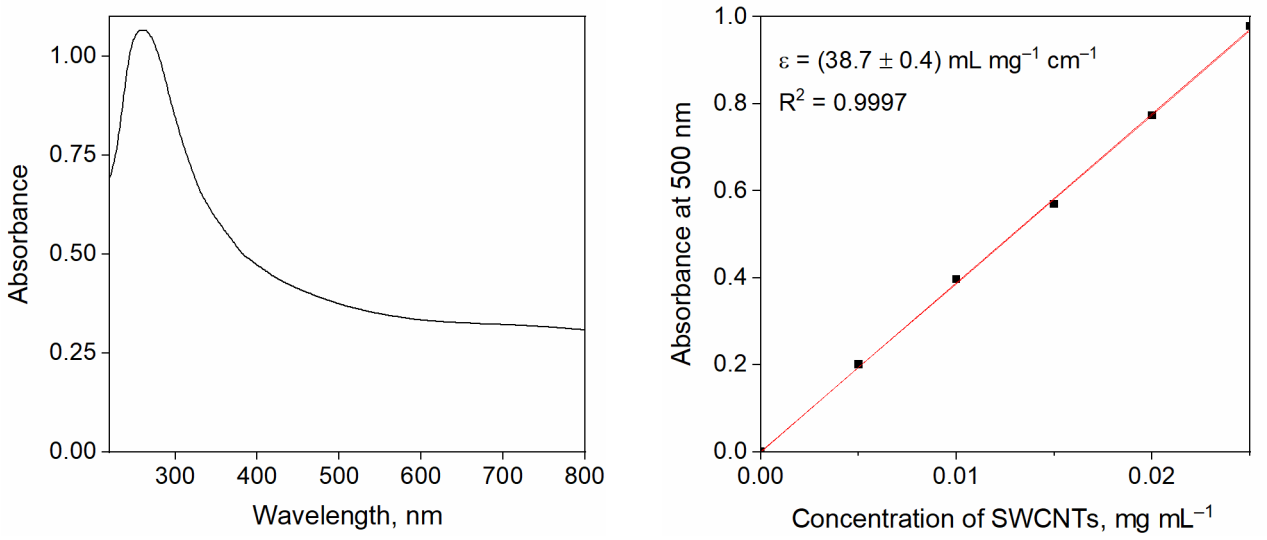

Figure S4 UV-vis. spectrum of SWCNT (0.39 mg mL^{-1}) in the aqueous HyPAN solution and the determination of extinction coefficient at 500 nm.

4. Galvanostatic profiles of the $LiFePO_4$-based electrodes prepared with different binders

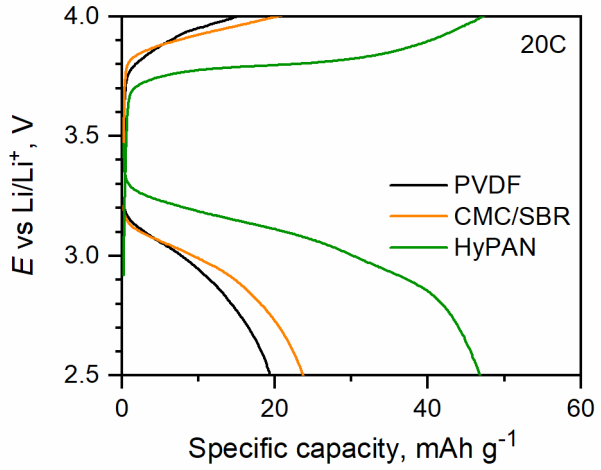

Figure S5 Galvanostatic profiles of the composite electrodes prepared with different polymer binders at 20C charge-discharge rate.

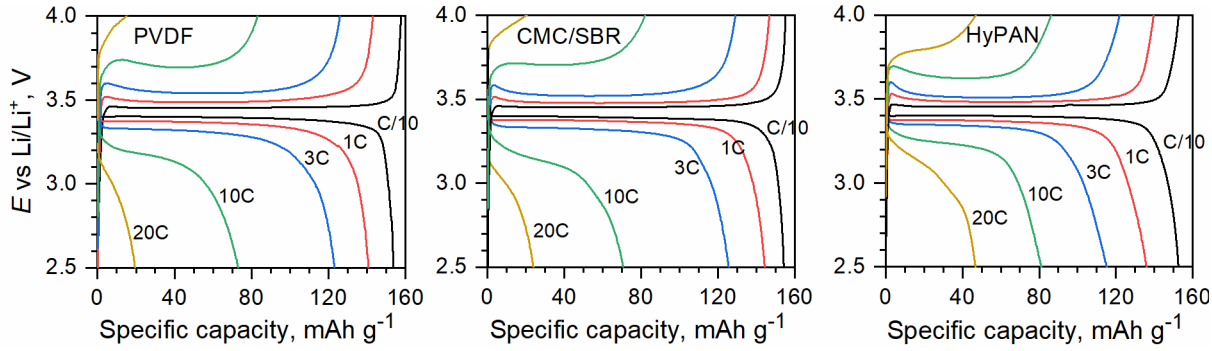

Figure S6 Galvanostatic charge-discharge curves of the composite electrodes prepared with different polymer binders.

Reference

S1 A. V. Kubarkov, A. A. Asharchuk, O. A. Drozhzhin, E. A. Karpushkin, K. J. Stevenson, E. V. Antipov and V. G. Sergeyev, *ACS Appl. Energy Mater.*, 2021, **4**, 12310; https://doi.org/10.1021/acsaem.1c02135.