Electronic Supplementary Information

Boosted adsorption-photocatalytic activities and potential lithium intercalation applications of layered potassium hexaniobate nano-family

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Futher details for preparation and characterization of the samples.

Preparation of KN-NL and KN-NL@PbS. The KN nanolaminas (KN-NL) were prepared by a mild hydrothermal process. Typically, 2.4 g Nb_2O_5 powder was added into 80 mL 0.9 mol/L KOH solution, the mixture was stirred for 1 h. The resulting mixed solvent was transferred into a 100 mL polyphenylene-lined stainless autoclave and hydrothermally treated at 200 °C for 15 h, followed by natural cooling to room temperature. The as-prepared products were obtained by centrifugation, washed several times with deionized (DI) water and absolute ethanol, and finally dried at 80 °C for 24 h in air. The resulting clear solution was collected for later use. The experimental procedures of KN-NL@PbS quantum dots can be described as follows, 0.2 g of the previously prepared KN-NL was added into 200 mL of DI water by ultrasonic dispersion for 20 min. Then it was stirred violently for 1h to ensure homogeneous suspension. Subsequently, an appropriate amount of 2-mercaptopropionic acid was added into the suspension. It refluxed at 65 °C for 1 h. Afterward, $Pb(CH_3COO)_2$ solution was added gradually drop by drop to the above reaction mixture under constant magnetic stirring at 65 °C for 1 h. Finally, Na_2S solution was added slowly to the system and stirred constantly at 65 °C for 1 h. The resulting product was extracted by centrifugation and washed repeatedly with DI water and absolute alcohol, then dried in a vacuum oven at 80 °C for 24 h. The pure PbS was prepared by following the above procedures only without adding KN-NL.

Preparation of KN-NS. For the synthesis of KN nano hollow spheres (KN-NS), 5 g of urea was added into 30 mL of the above clear solution. This mixed clear solution was transferred into a 50 mL polyphenylene-lined stainless autoclave and hydrothermally treated at 200 °C for 15 h. The final products were collected by centrifugation, washed with deionized water and absolute ethanol for several times, and then dried at 80 °C for 24 h in air.

Preparation of (K, Nb)-precursors. In this work, a novel method was proposed to prepare the (K, Nb)-precursors, which is crucial for the synthesis of KN nanocrystalline films. This method is based on the hydrothermal process, the experimental procedures can be described as follows. 0.2 g of the white precipitate which was obtained by adding absolute ethyl alcohol into the above clear solution, with 15 mL ethanediol, were stirred for 1 h. Then the mixed solution was transferred

into a 50 mL polyphenylene-lined stainless autoclave and hydrothermally treated at 200 °C for 20 h. The yellow clear solution was obtained after natural cooling to room temperature. Finally the hydrothermal derived (K, Nb)-precursor (HT-precursor) was obtained by mixing the above yellow clear solution and acetic acid (CH_3COOH) with a volume ratio of 1:1.

Preparation of KN film. To obtain KN films, the HT-precursor solutions were spin-coated onto Si substrate. The films were deposited by employing a spin-coating process at a speed of 4000 rpm for 20 s. Each layer of the films was dried at 200 °C for 200 s to evaporate the solvent and then pyrolyzed at 400 °C for 200 s to remove residual organic compounds, followed by annealing at 680 °C for 300 s to crystallize the films by a rapid thermal annealing procedure. The spin-coating and annealing-treatment procedures were repeated several times to obtain a desired thickness.

Characterization methods. The crystal phase structures of all samples were analyzed by X-ray diffraction (XRD, Cu Kα, D8 Advance, Bruker). Raman scattering experiments were carried out by a micro-Raman spectrometer (Jobin-Yvon LabRAM HR 800UV). The surface morphologies of powder samples were examined by field emission scanning electron microscopy (FESEM: Philips XL30FEG). The transmission electron microscopy (TEM) images, selected area electron diffraction (SAED), and chemical compositions of the as-synthesized composites were acquired by TEM (JEM 2100F, Japan) with an acceleration voltage of 200 kV, equipped with an energy dispersive X-ray spectroscopy (EDS) detector for elemental mapping. Ultraviolet-visible light diffuse reflectance spectra (UV-VIS DRS) were recorded by a double beam infrared-ultraviolet spectrometer (Perkin-Elmer UV/VIS Lambda 950) equipped with an integrating sphere assembly. The Brunauer-Emmett-Teller (BET) surface areas of the powder samples were measured by a surface area analyzer (TriStar I-I 3020), and the pore-size distribution curves were obtained using Barrett-Joiner-Halenda (BJH) model. Thermogravimetric analyses (TGA) and differential scanning calorimetry (DSC) of samples were performed in a TGA/DSC 1 STAR^e System (Mettler-Toledo) from 30 °C to 1000 °C with a heating rate of 20 °C/min.

Electrochemical tests. Cyclic voltammetry (CV) was implemented on a CHI660E electrochemical workstation with a three-electrode system in an aqueous solution of 0.1 mol/L HCl at a

scan rate of 100 mV/s. The as-prepared KN-NL and KN-NS were used as a working electrode, a platinum wire as the counter electrode and Ag/AgCl as the reference electrode. The working electrodes were prepared by adding appropriate amount of a slurry, which consisted of 80 wt% active materials, 10 wt% acetylene black and 10 wt% polyvinylidene fluoride (PVDF) dissolved in N-methyl-2-pyrrolidinone (NMP), then dried in a vacuum oven at 90 °C for 20 h. Electrochemical impedance spectroscopy (EIS) were performed by using an impedance measurement unit of the electrochemical workstation in the frequency range of 0.1-10^6 Hz, with an ac amplitude of 5 mV.

Photocatalytic degradation of dyes. The catalytic performance of different samples were evaluated by observing their abilities to adsorb (in dark) and degrade (under light irradiation) the Rhodamine B (RhB) and methylene blue (MB) dye. The UV-light source was a 500 W long arc Hg lamp equipped with a filter which only allows UV-light through. The visible-light source was a 500 W Xe lamp equipped with a filter which only allows visible-light through. The photocatalytic experiments were carried out by a reactor equipped with a cooling water cycle system, which can simultaneously conduct six parallel reactions. In a typical photocatalytic test, the catalyst (50 mg) was dispersed in a 50 mL aqueous solution of RhB dye (10-20 mg/L) with different pH (3-11), then the mixed suspensions were magnetically stirred in the dark at room temperature for 1h to establish adsorption-desorption equilibrium. After light irradiation, adequate volume of the suspension were extracted and centrifuged at an interval of 10/15 minutes for analysis. Aqueous solution of MB dye (50 mg/L) with different volume (20-100 mL) and different pH (3-11) were used to perform the adsorption experiments. Note that the KOH and HCl aqueous solution were used to adjust the pH of the mixed solution with the aid of a pH meter (Mettler-Toledo). The adsorption and photocatalysis efficiency were investigated by measuring the change in intensity of the characteristic absorbance of RhB and MB dye using spectrometer (Perkin-Elmer UV/VIS Lambda 950). The mineralization rate of the solution was analyzed using the total organic carbon (TOC) analyser (Shimadzu, TOC-L CPN).

Lithium ion batteries tests Two-electrode coin-cell (CR-2025) assembled in an argon-filled glovebox were used for the energy storage investigation. The KN-NL was use as the working elec-

trode, which was fabricated by casting a slurry of 80 wt% active material, 10 wt% acetylene black, and 10 wt% polyvinylidene fluoride in N-methyl-2-pyrrolidinone (NMP) on a copper foil, then dried at 90 °C in vacuum for 20 h. The lithium foil was used as counter and reference electrodes, a membrane (Celgard 2400) as the separator, and the electrolytes were 1 mol/L $LiPF_6$ solution in a 1:1:1 (by volume) mixture of ethylene carbonate (EC), dimethyl carbonate (DMC) and ethyl methyl carbonate (EMC). Galvanostatic charge-discharge experiments were performed at different current densities in the voltage range of 0.02-3.0 V (*vs.* Li^+/Li) using battery measurement system (LAND-CT2001A). Cyclic voltammetry (CV) at a sweep rate of 0.5 mV/s, and electrochemical impedance spectroscopy (EIS) with an amplitude of 5 mV in the frequency range 100 kHz to 0.01 Hz, were performed on an electrochemical workstation (CHI660E). All electrochemical measurements were carried out at room temperature.

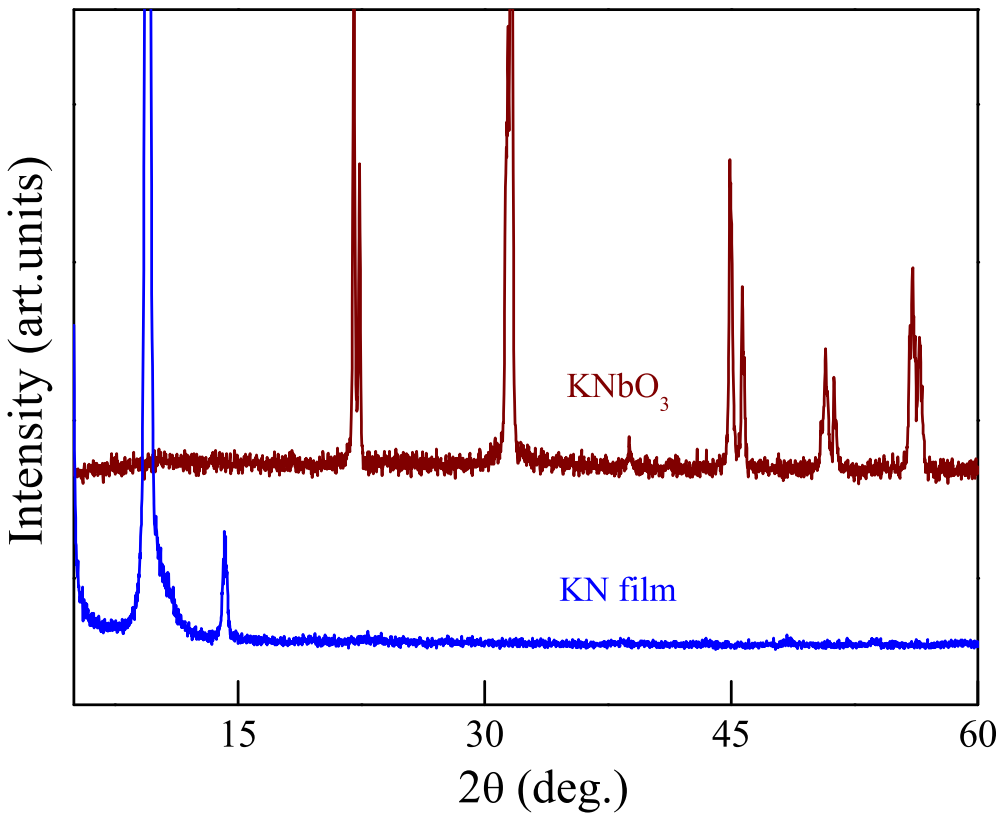

Fig. S 1: XRD patterns of KN film and $KNbO_3$ which was obtained by calcining the white precipitate at 800 °C for 4 h.

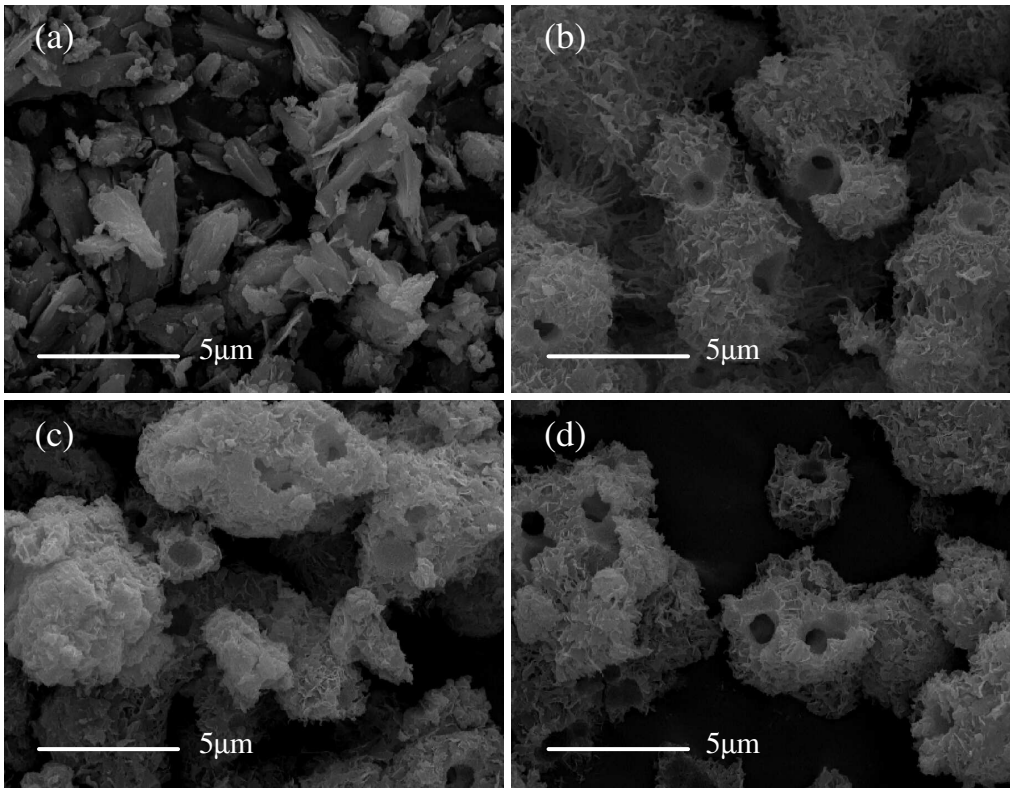

Fig. S 2: SEM images of KN-NS at different hydrothermal condition: 1 g (a) and 3 g (b) of urea at 200 °C for 15 h, 5 g of urea at 200 °C for 5 h (c) and 10 h (d).

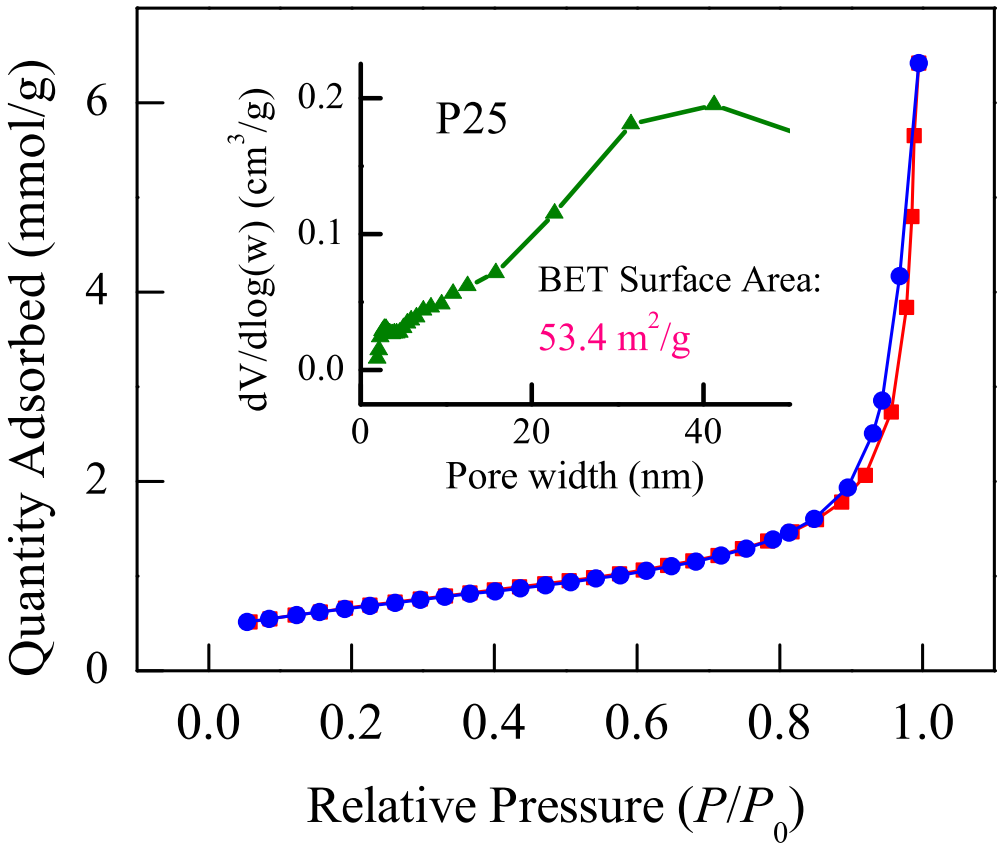

Fig. S 3: Nitrogen adsorption-desorption isotherm and pore-size distribution (inset) for P25 TiO_2.

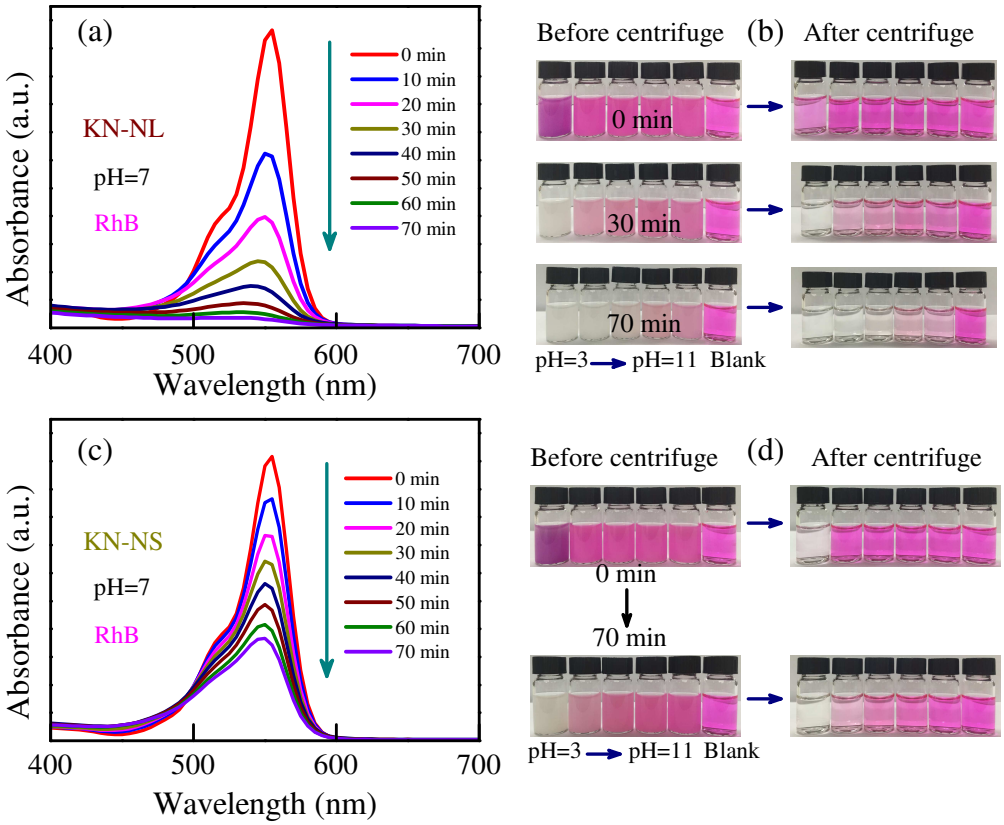

Fig. S 4: The corresponding adsorption spectra of an aqueous solution of RhB for (a) KN-NL and (c) KN-NS at the irradiation time of 0-70 min when pH=7. The color contrast of the relevant RhB dye solution for (b) KN-NL and (d) KN-NS.

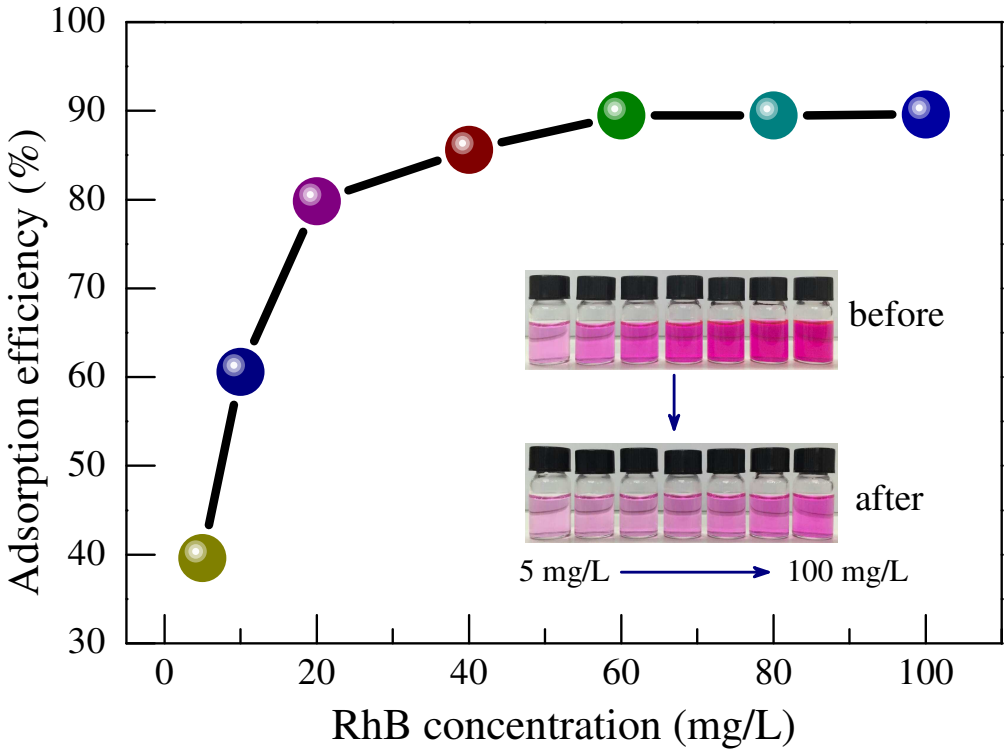

Fig. S 5: The adsorption efficiency for KN-NL in the RhB solution with differernt concentrations (5, 10, 20, 40, 60, 80, 100 mg/L, pH=3). Note that the inset is the relevant color contrast.

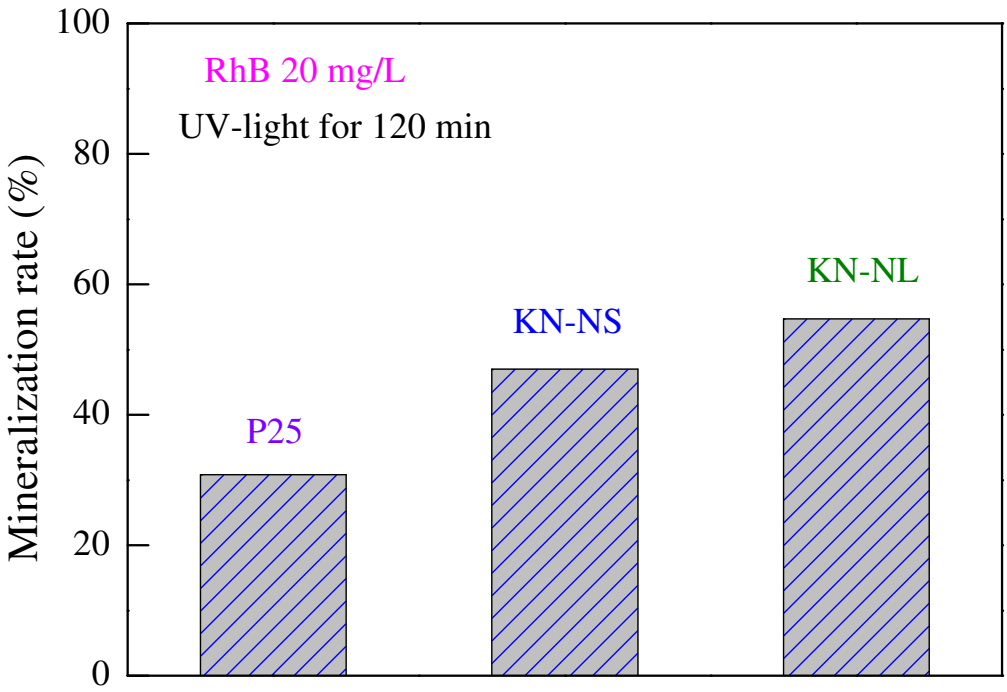

Fig. S 6: Photocatalytic mineralization rates of RhB solution for P25, KN-NS and KN-NL under UV-light irradiation.

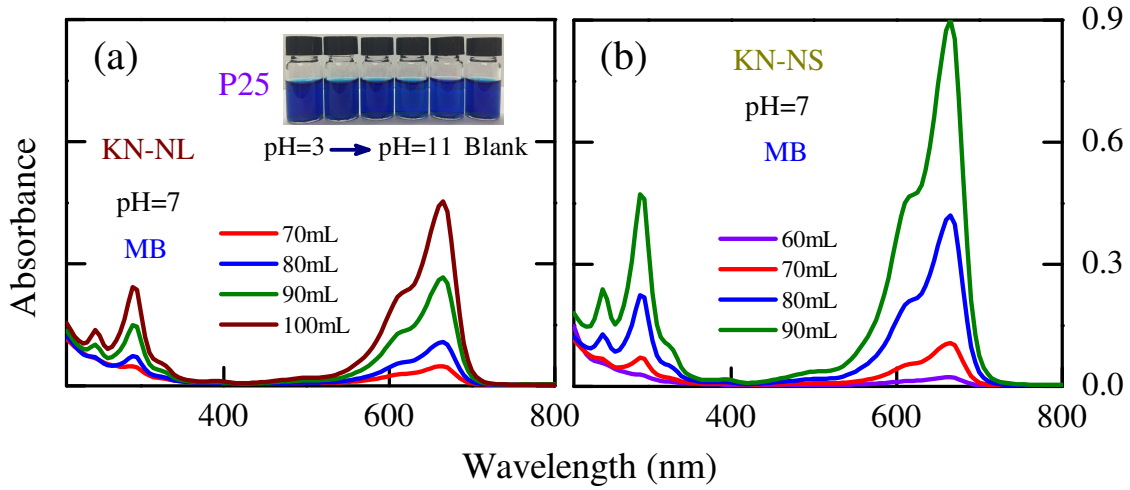

Fig. S 7: The corresponding adsorption spectra of an aqueous solution of MB for (a) KN-NL and (b) KN-NS at different adsorption capacity of 60-100 mL when pH=7. Note that the inset is the color contrast of MB dye adsorption in the pH range of 3-11 for P25.

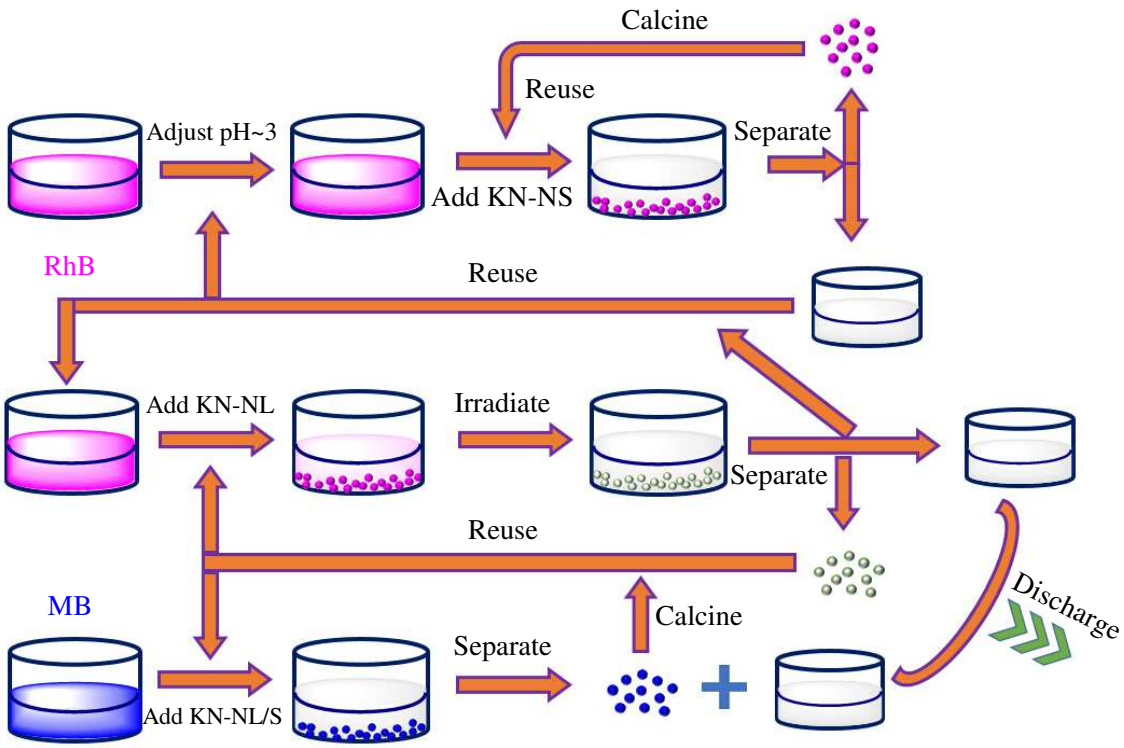

Fig. S 8: A high-efficient scheme to deal with the dye wastewater.

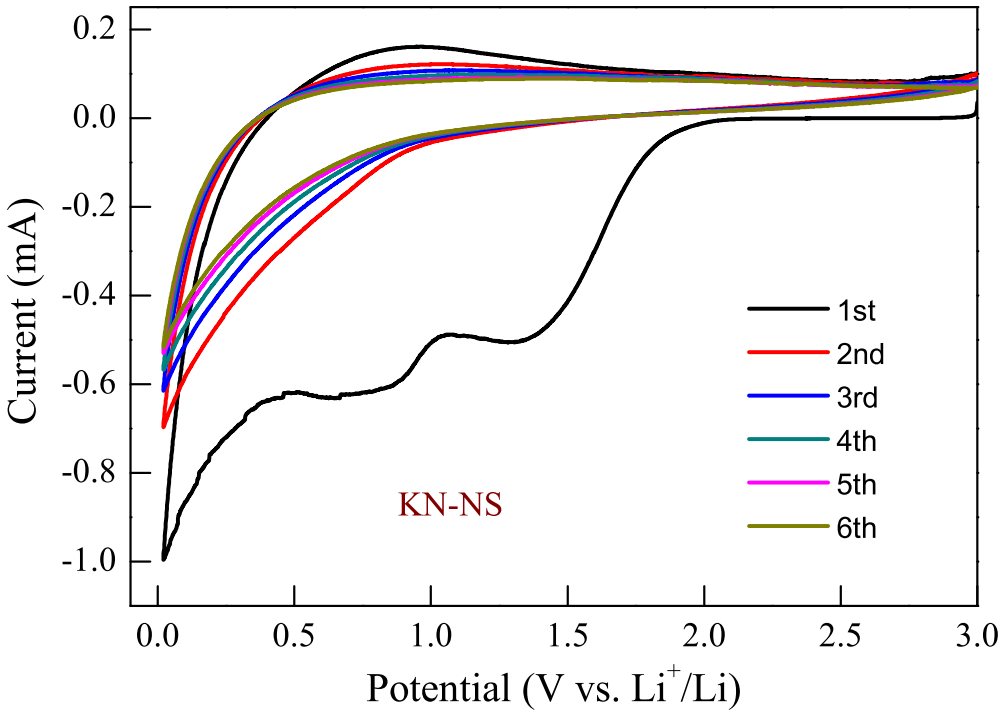

Fig. S 9: Cyclic voltammetry (CV) curves of KN-NS measured at a scan rate of 0.5 mV/s in the voltage range of 0.02-3.0 V (*vs.* Li^+/Li).

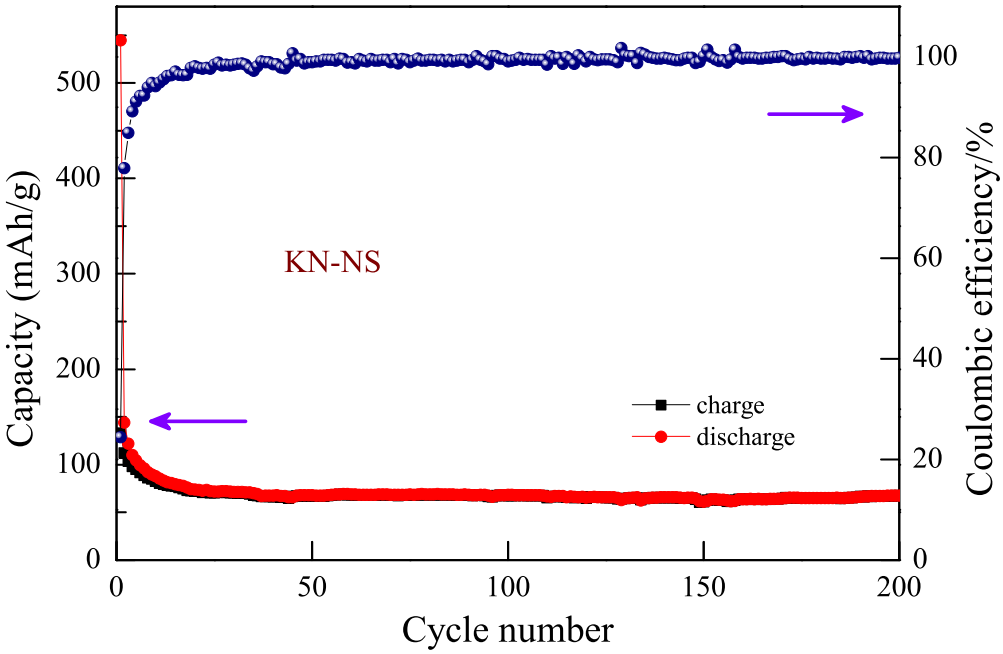

Fig. S 10: Cycling stability and coulombic efficiency of KN-NS at a current density of 200 mA/g.