Electronic Supplementary Material

Hierarchically porous cellulose nanofibril aerogel decorated with polypyrrole and nickel-cobalt layered double hydroxide for high-performance nonenzymatic glucose sensors

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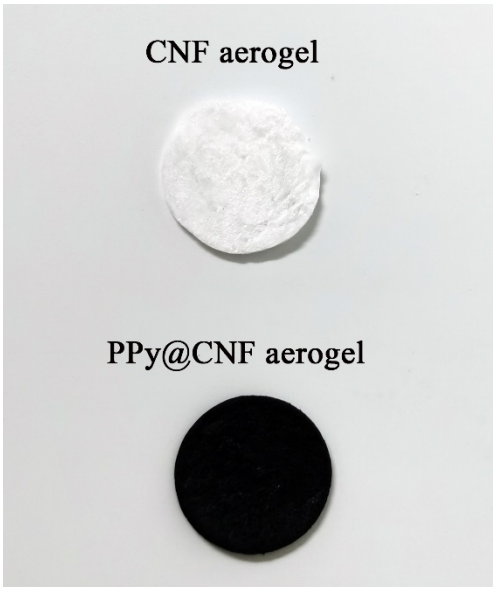

Figure S1. Digital photographs of the cellulose nanofibril (CNF) aerogel and PPy@CNF aerogel.

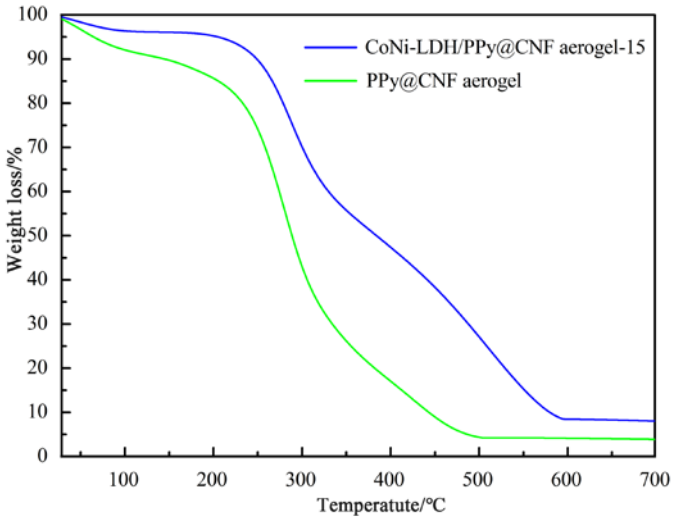

Figure S2. TG curves of the CoNi–LDH/PPy@CNF aerogel-15 and PPy@CNF aerogel.

The residue contents of the CoNi–LDH/PPy@CNF aerogel-15 and PPy@CNF aerogel are 8.02% and 3.89% at 700 °C, respectively. The primary residue at 700 °C possibly includes char, CoO, and NiO for the CoNi–LDH/PPy@CNF aerogel-15. Therefore, the proportion of CoNi–LDH in the CoNi–LDH/PPy@CNF aerogel-15 is (8.02% – 3.89%)/75*93 = 5.1 wt.%.

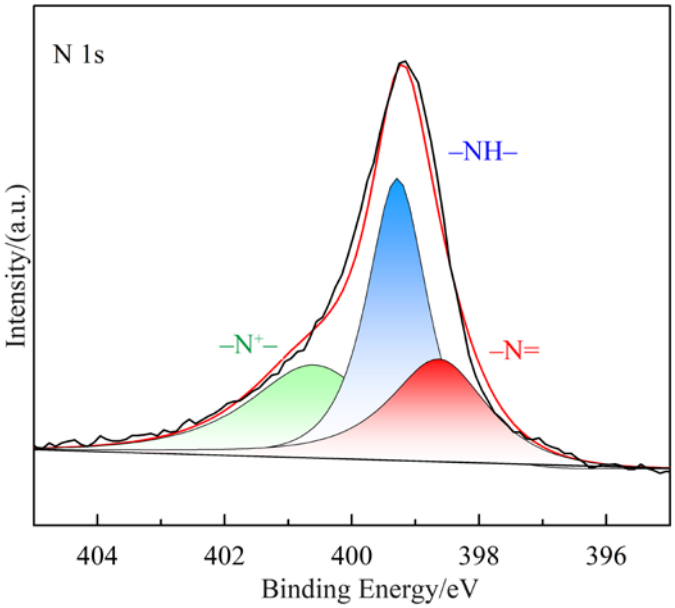

Figure S3. N 1s core-level X-ray photoelectron spectroscopy (XPS) spectrum of the PPy@CNF aerogel.

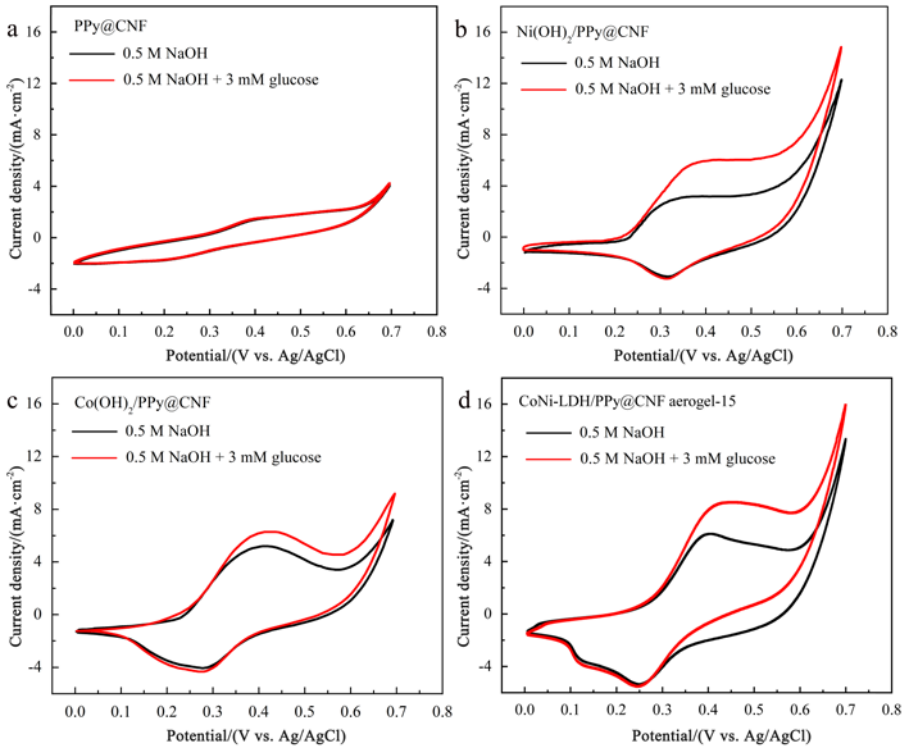

Figure S4. Cyclic voltammetry (CV) curves of the PPy@CNF aerogel (a), $Ni(OH)_2$/PPy@CNF

aerogel (b), $Co(OH)_2$/PPy@CNF aerogel (c), and CoNi–LDH/PPy@CNF aerogel-15 (d) with and without 3.0 mM glucose at a scan rate of 20 $mV \cdot s^{-1}$.

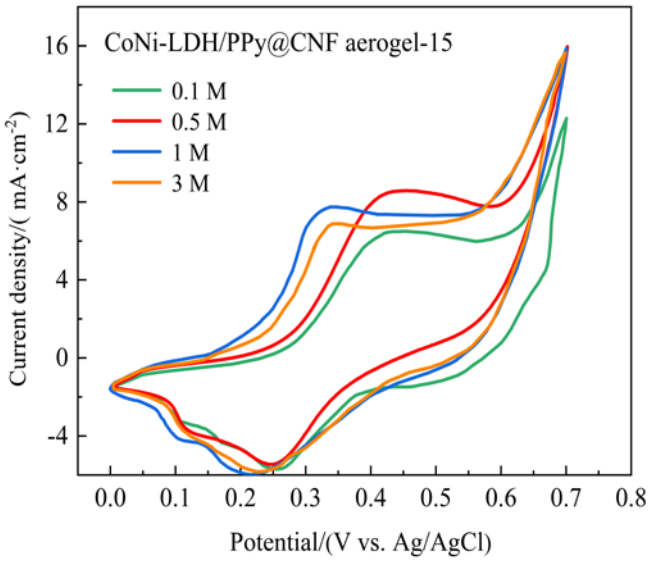

Figure S5. CV plots of the CoNi–LDH/PPy@CNF aerogel-15 tested in various concentrations of NaOH solution with 3 mM glucose at 20 $mV \cdot s^{-1}$.

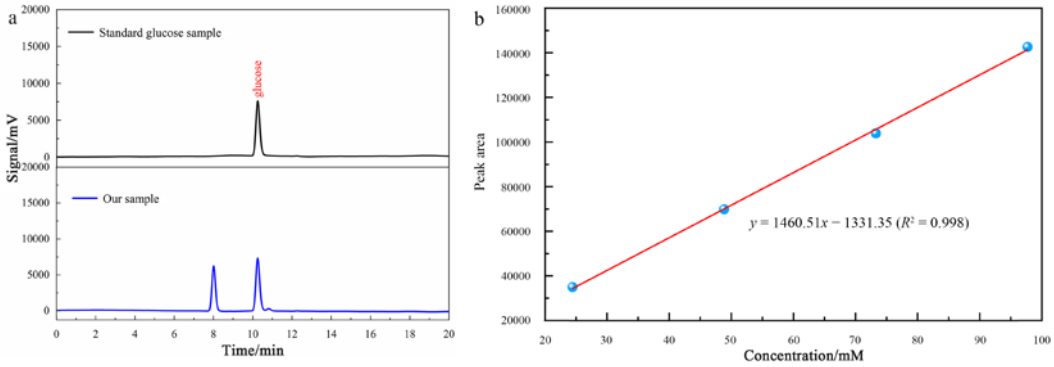

Figure S6. Chromatogram after determination of glucose concentration by high-performance liquid chromatography (HPLC) and the standard curve graph. (a) HPLC chromatograms. (b) Calibration curve of glucose measurement using an HPLC method.

In this study, the CoNi–LDH/PPy@CNF aerogel-15 electrode is immersed in a mixed solution of 0.5 M NaOH and 75 mM glucose (40 mL); subsequently, a CV test (half of one turn) is conducted over a voltage range of 0 to 0.7 V (vs. Ag/AgCl) at a scanning rate of 20 $mV \cdot s^{-1}$. To ensure accurate results within the detectable range of HPLC, a glucose concentration of 75 mM is chosen, which is slightly above the minimum detectable concentration of 70 mM. HPLC analysis is conducted using an autosampler at a flow rate of 0.5 mL min^{-1} with a mobile phase consisting of 75% acetonitrile and 25% sterile ultrapure water. Each sample is injected through a guard column with a 20-μL volume, followed by separation on an NH_2 column at 35 °C. The RIA-10A refractive index detector is used with an evaporator temperature of 60 ℃ and a nebulizer temperature of 40 ℃. The glucose concentrations are studied based on the signal of each sample at corresponding retention times. Figure S6 illustrates the chromatogram and standard curve diagram resulting from the HPLC determination of glucose concentration. As shown in Fig. S6a, the peak position at approximately 10.3 minutes in the chromatogram of the sample being analyzed is consistent with that in the chromatogram of the standard glucose sample. Thus, we infer that the tested sample contains glucose. Various concentrations of standard solutions are prepared, and the glucose calibration curve is obtained using the HPLC method. Fig. S6b shows a good linear relationship between the peak area and glucose concentration in the range of 25~95 mM, and the regression equation is $y = 1460.51x - 1331.35$ (R^2 = 0.998). According to this equation, the glucose concentration after the CV test is

74.84 mM according to the peak area of the sample solution (y = 107969). Thus, the conversion content of glucose following the CV test (half a turn) is determined by dividing the difference in concentration before and after the test by the weight of the active material (CoNi–LDH) in the CoNi–LDH/PPy@CNF aerogel-15 electrode. The conversion content is (75 mM − 74.84 mM) × 40 mL ÷ (12.9 mg × 5.1 wt.%) = 9.7 × 10^{-3} mol/mg. The weight of the CoNi–LDH/PPy@CNF aerogel-15 electrode with a size of 1 cm × 1 cm × 0.1 cm is 12.9 mg, and the proportion of CoNi–LDH in the CoNi–LDH/PPy@CNF aerogel-15 is 5.1 wt.% according to the TG test (Fig. S2).

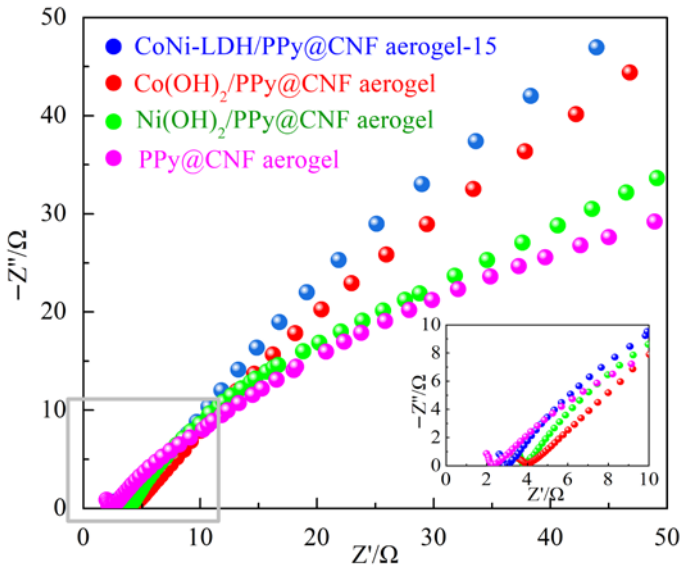

Figure S7. Nyquist plots of the PPy@CNF aerogel, CoNi–LDH/PPy@CNF aerogel-15, $Ni(OH)_2$/PPy@CNF aerogel, and $Co(OH)_2$/PPy@CNF aerogel. (The inset shows the enlarged image at high frequency).

Table S1. Known or potential interferents and their general levels in human blood.

Interferents	**General level (mg/dL)**	**Data sources**
Ascorbic acid	0.4~2	Test ID: VITC Ascorbic Acid (Vitamin C), Plasma, provided by Mayo Clinic Laboratories https://www.mayocliniclabs.com/test-catalog/Clinical+and+Interpretive/42362
Dopamine	$0\sim30\times10^{-7}$	Medical Encyclopedia, provided by U.S. National Library of Medicine https://medlineplus.gov/ency/article/003561.htm
Uric acid	2.0~6.5	Maiuolo J, Oppedisano F, Gratteri S, Muscoli C. Regulation of uric acid metabolism and excretion. International Journal of Cardiology, 2016, 213: 8-14
Fructose	0.12~0.16	Takahiro K, Hiroshi A, Toshikazu Y. Increased fructose concentrations in blood and urine in patients with diabetes. Diabetes Care, 2002; 25 (2): 353–357
Lactose	<0.5	Lorenz C, Sandoval W, Mortellaro M. Interference assessment of various endogenous and exogenous substances on the performance of the eversense long-term implantable continuous glucose monitoring system. Diabetes Technology & Therapeutics, 2018, 20(5): 344-352
Sucrose	0.082~0.29	Shishido T, Yamaguchi T, Odaka T, Seimiya M, Saisho H, Nomura F. Significance of a novel sucrose permeability test using serum in the diagnosis of early gastric cancer. World Journal of Gastroenterology, 2005, 11(44): 6905
Folic acid	$4.17\sim20\times10^{-4}$	Galukande M, Jombwe J, Fualal J, Baingana R, Gakwaya A. Reference values for serum levels of folic acid and vitamin B12 in a young adult Ugandan population. African Health Sciences, 2011, 11(2): 240-243