Supporting Information

Thin Films Based on Nanocomposites Between Crumpled Graphene Fully Decorated by Prussian Blue: A New Material for Aqueous Battery System

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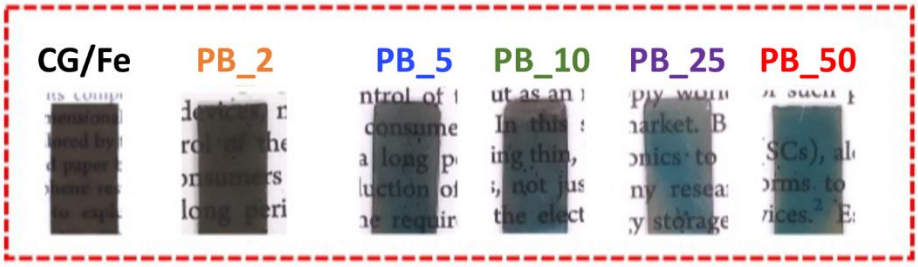

Figure S1: Photographs of the films based on CG/PB composites.

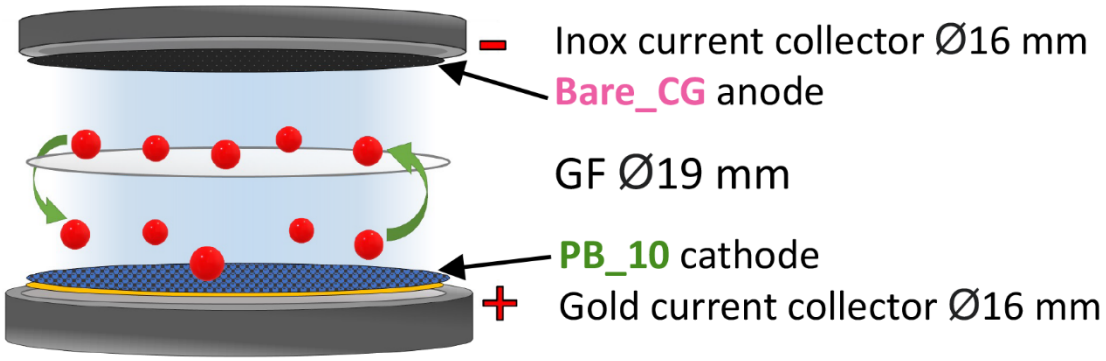

Figure S2: Schematic representation of the assembled coin cell (CR2032) devices.

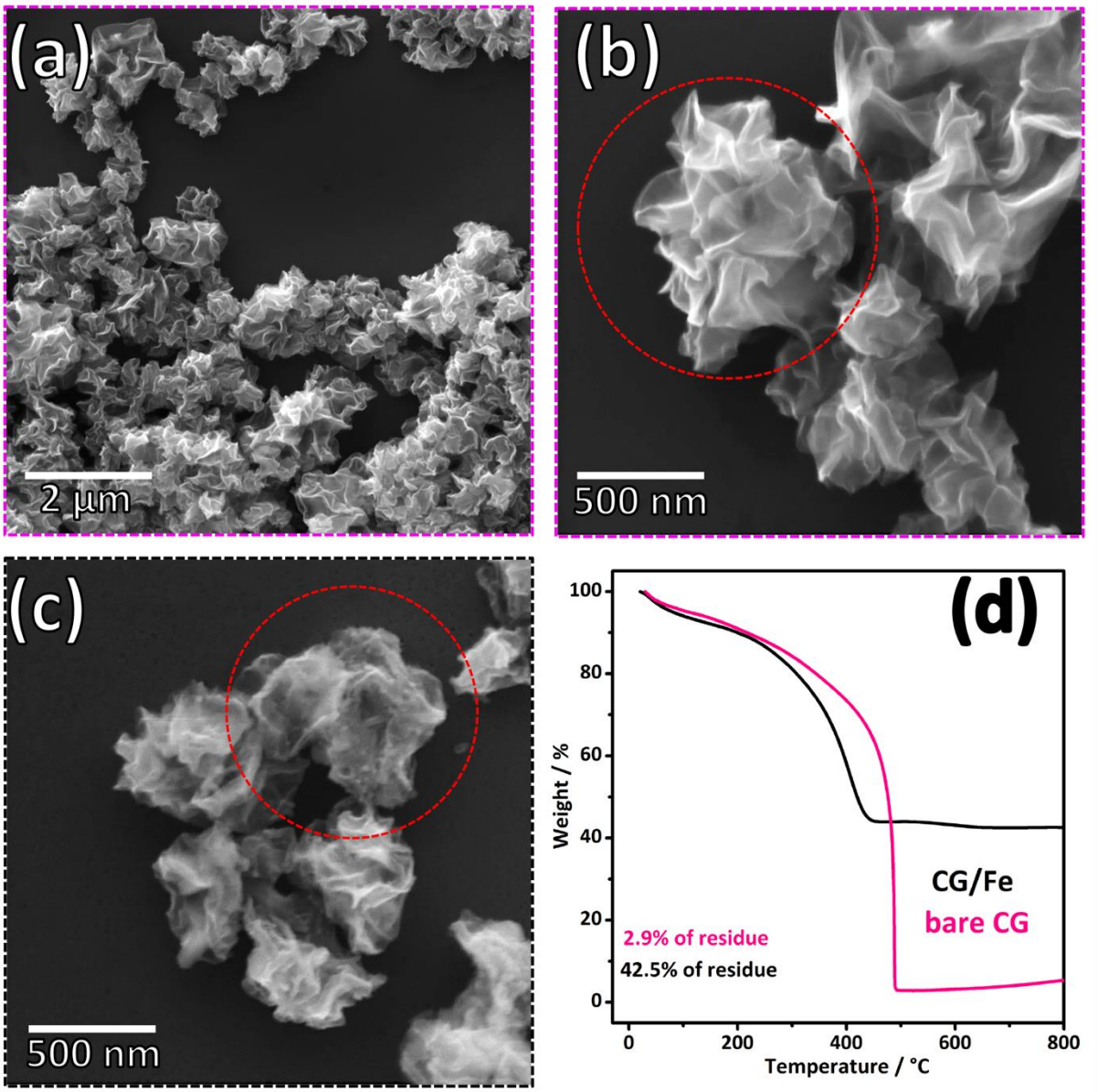

Figure S3: Bare_CG sample SEM images (a,b), CG/Fe sample (c), and thermogravimetric analyses (d).

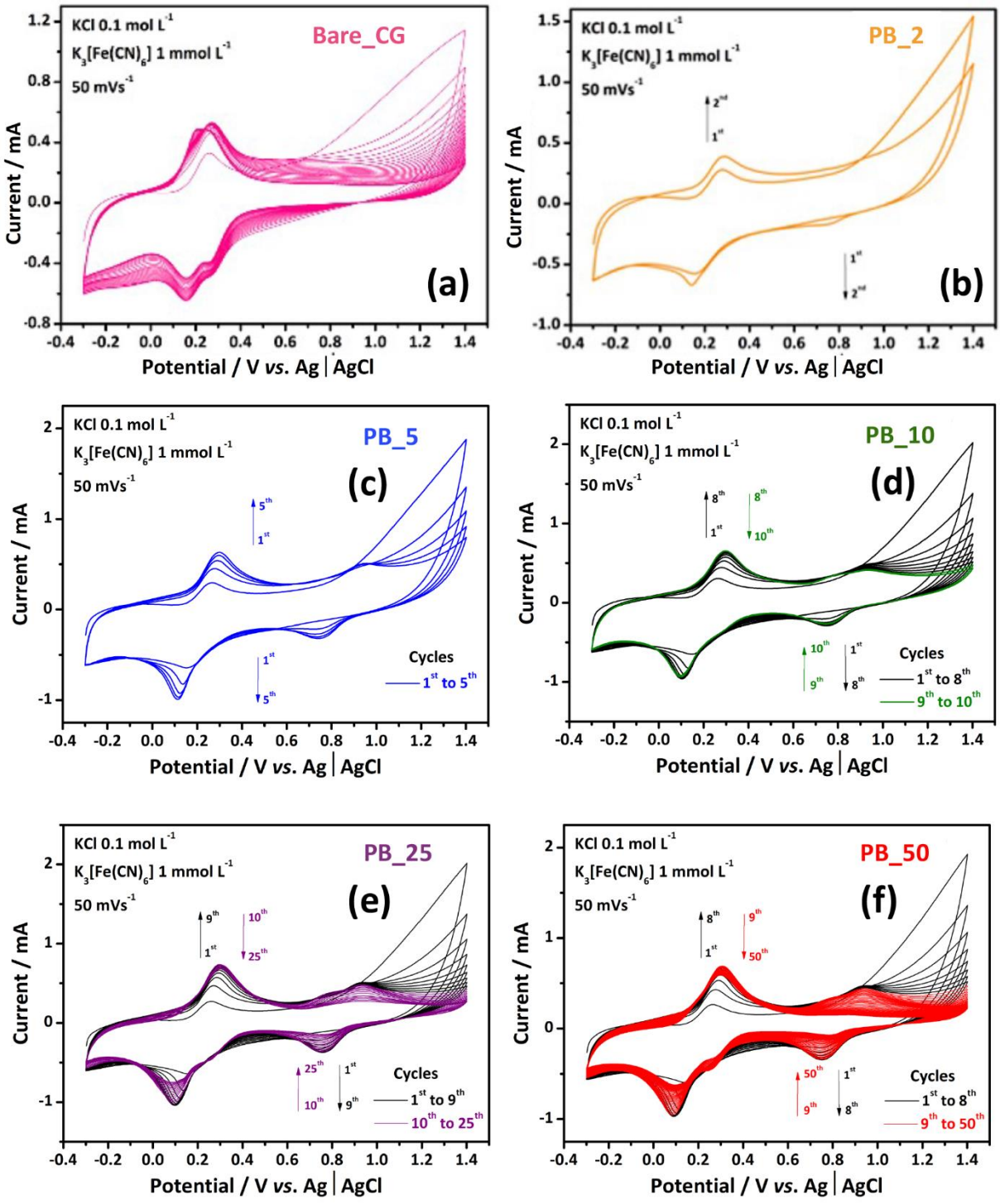

Figure S4: Electrodeposition of PB nanocubes performed in 50 cycles, with a scan rate of 50 mV s^{-1}, 0.1 mol L^{-1} KCl electrolyte, and 0.1 mmol L^{-1} $K_3[Fe(CN)_6]$, for all samples.

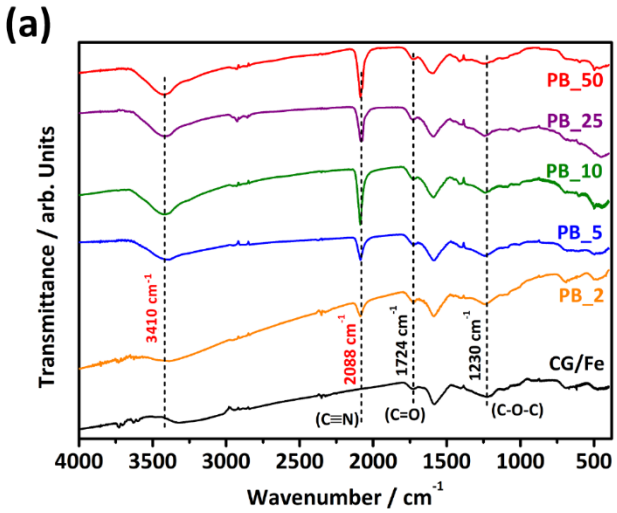

Figure S5: FTIR spectra of the samples CG/Fe, PB_2, PB_5 PB_10, PB_25, and PB_50.

The infrared spectra of the CG/Fe sample and PB_2, PB_5, PB_10, PB_25, and PB_50 composites are depicted in Figure S5. The three main bands observed in the CG/Fe sample, at 1230, 1724, and 3320 cm^{-1} correspond to the epoxides group, carboxylic acids stretching and hydroxyls stretching, respectively. As shown in the TGA analysis in Figure S3 (d), these bands originate from oxygenate functional groups remaining from GO, indicating the presence of these groups in CG samples. [1,2] A different behavior was observed for the composites decorated with Prussian blue, where the cyano ligand band at 2088 cm^{-1} proves the presence of PB in the samples. Although the Raman spectra well evaluated PB, the FTIR analysis showed the presence of a band at 3410 cm^{-1} for the composites with PB referring to OH-stretching and the possible presence of interstitial water.[3]

[1] Santos, Y. H.; Hostert, L.; Almeida, T. S. D.; Zarbin, A. J. G.; Souza, V. H. R. and Orth, E. S., *Journal of the Brazilian Chemical Society*, 2025, 35, e20240063.

[2] Acik, M.; Lee, G.; Mattevi, C.; Pirkle, A.; Wallace, R. M.; Chhowalla, M.; Cho, K. and Chabal y., *The Journal of Physical Chemistry C*, 2011, 115, 19761-19781.

[3] Singh, S.; Pandey, P. C., *Journal of Environmental Chemical Engineering*, 2020, 8, 103753.

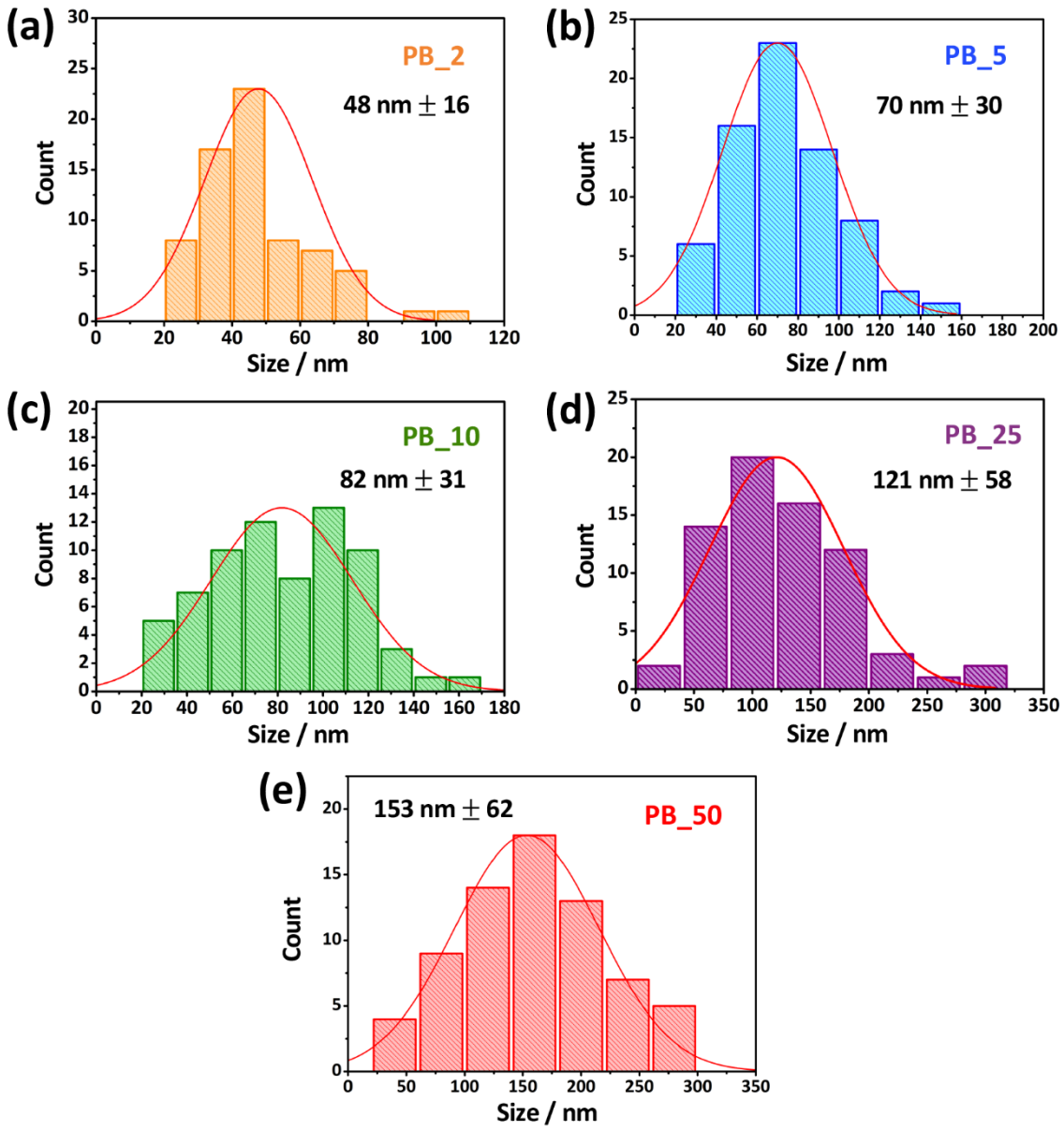

Figure S6: Size distribution of PB nanoparticles in samples PB_2 (a), PB_5 (b), PB_10 (c), PB_25 (d), and PB_50 (d).

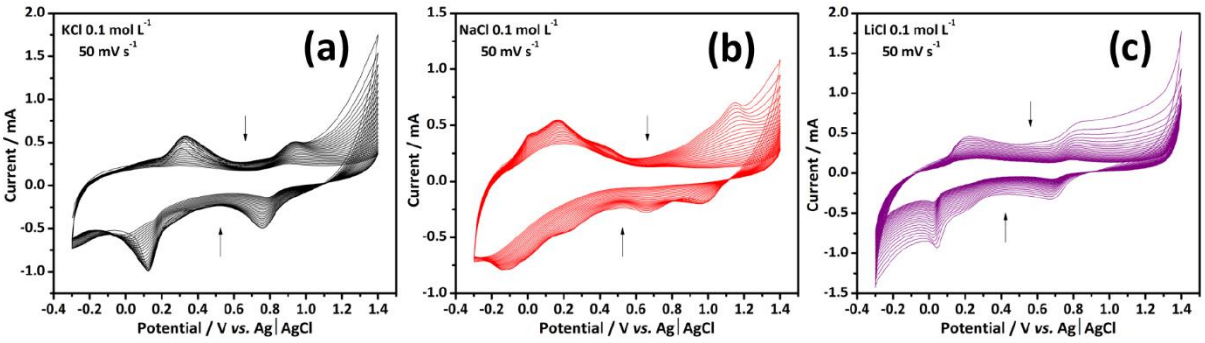

Figure S7: Voltammetric profiles stability of the PB_10 electrode in KCl 0.1 mol L^{-1} (a), NaCl 0.1 mol L^{-1} (b), and LiCl 0.1 mol L^{-1} (c) over 25 CV cycles, 50 mV s^{-1}, -0.3 to 1.4 V, pH 5.0.

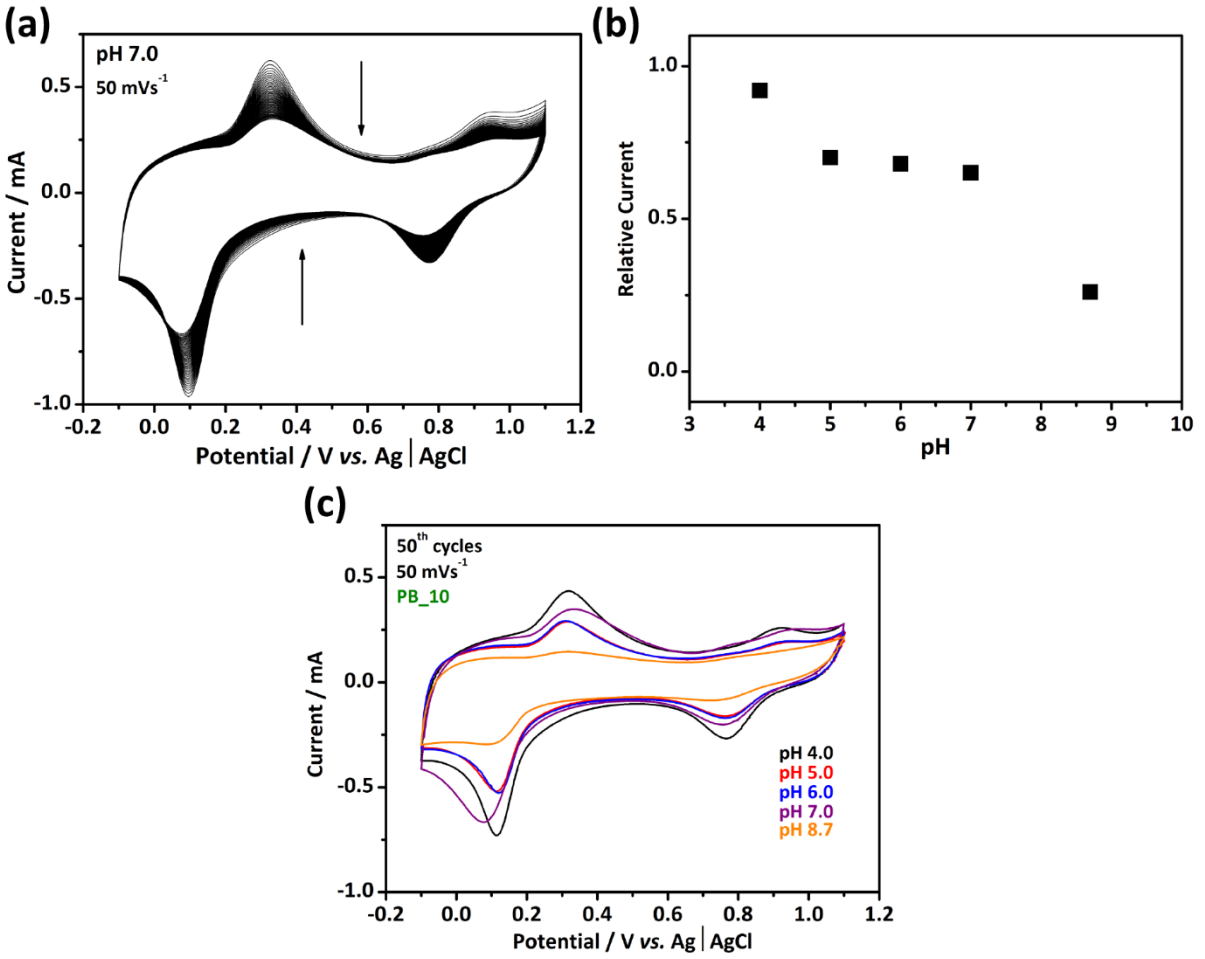

Figure S8: Voltammetric profile stability of the PB_10 electrode in KCl 0.1 mol L^{-1}, pH 7.0, over 50 CV, 50 mV s^{-1}, -0.1 to 1.1 V (a). Curve of relative current intensity between the 2nd and 50th CV for the cathodic peak located between 0.0 and 0.2 V (b) and voltammetric profiles in differents pH values (c) after 50 cycles of stability analysis.

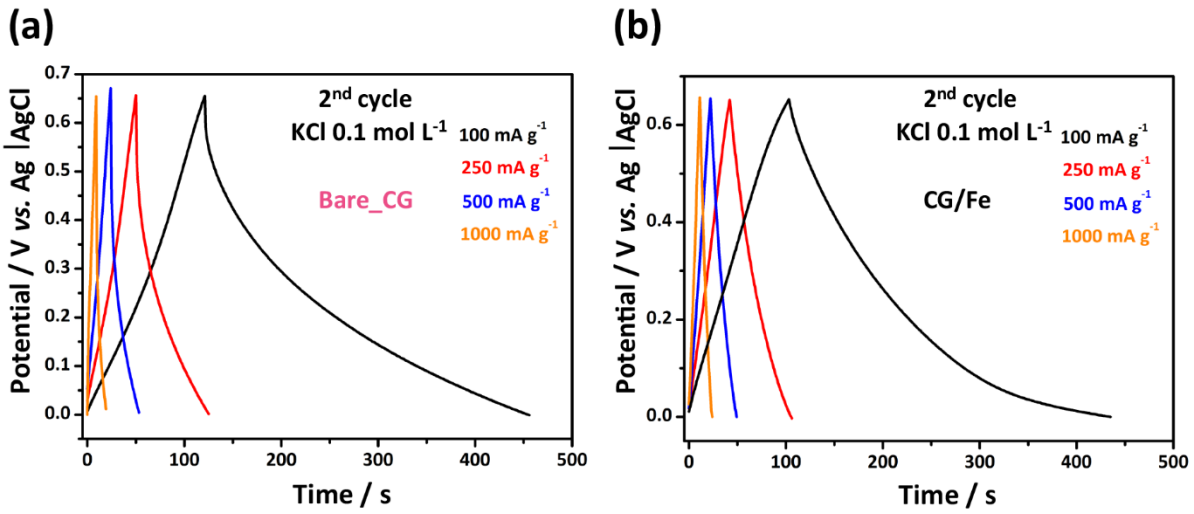

Figure S9: Charge/Discharge profile for Bare_CG (a) and CG/Fe (b) samples, in KCl 0.1 mol L^{-1}, 0.0 to 0.65 V

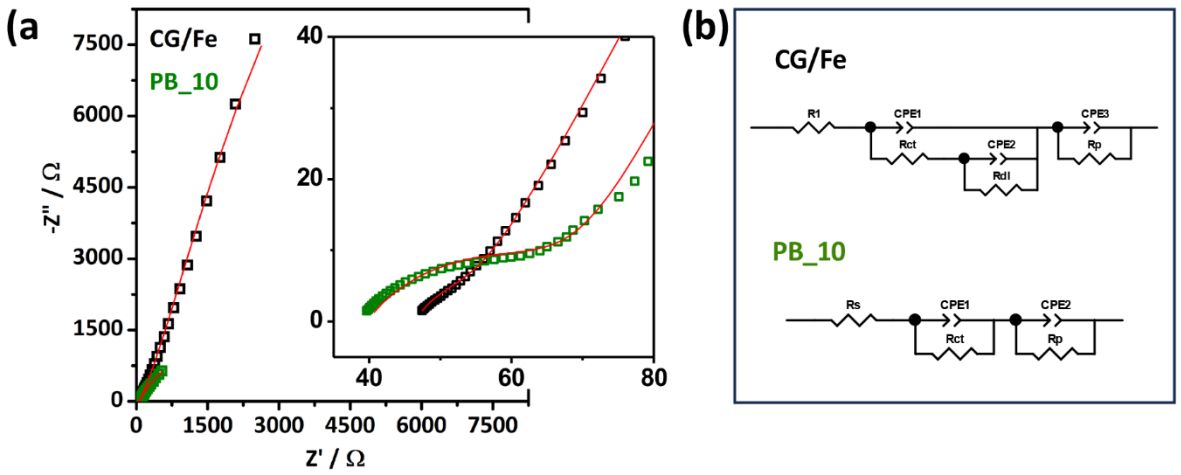

Figure S10: Electrochemical impedance spectroscopy (EIS) profiles for CG/Fe and PB_10 electrodes (a) and equivalent circuits for EIS fitting in (b).

Table S1: Atomic percentage, XPS peak positions, and elements of the samples CG/Fe and PB_10.

Samples	Atomic percentage / %						
	C (1s)	O (1s)	Fe ($2p_{3/2}$)	N (1s)	Cl (2p)	S (2p)	K (2p)
CG/Fe	48.01	30.96	11.35	2.33	5.71	1.64	---
PB_10	48.43	33.15	7.93	9.63	---	---	0.86

Element	Peak position/ eV		Assignment
C (1s)	285.0	284.9	C=C arom.
	---	285.4	Fe^{II}–CN
	286.6	286.4	C–O
	288.3	288.7	C=O
	289.5	289.8	O–C=O
Fe ($2p_{3/2}$)	---	708.7	Fe^{II}–CN
	710.8	710.4	Fe_2O_3
	712.3	712.0	FeOOH
	714.0	713.9	Fe^{III} satellite

Table S2: Specific capacities of the CG/PB electrodes at different current densities.

	Current Density / mA g^{-1}					
Sample	**250**	**350**	**500**	**750**	**1000**	**2000**
PB_2	29.7	26.4	23.1	20.8	29.4	28.3
PB_5	51.0	42.0	36.3	30.0	31.2	25.3
PB_10	62.7	56.6	50.4	45.2	43.9	40.0
PB_25	58.7	49.7	41.8	35.6	33.9	29.4
PB_50	50.0	45.5	39.6	37.5	35.5	30.0