

Supporting Information:

Synergistic effect of novel redox additives of *p*-nitroaniline and dimethylglyoxime for highly improving the supercapacitor performances

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Optical analysis:

Fourier transform infrared (FT-IR) spectroscopy of the **carbon-blank** and **carbon-DMG-0.15** was performed by a Nicolet 67 spectrometer (Thermo Nicolet, American) at room temperature (RT) for mid-IR (the frequency range of $4000 \sim 400$ cm^{-1}) data collection. Room-temperature UV-vis spectroscopy was recorded between the range of $200 \sim 800$ nm by a UV-vis spectrophotometer (UV-2550, Shimadzu Corporation, Japan) and the UV-vis absorption experiments were carried out on the 1 cm path length spectrometric quartz cell.

Table S1. Summary of DMG and PNA conditions in 6 mol L⁻¹ KOH electrolyte.

NO.	Samples (abbreviation)	DMG	PNA	KOH
		(mmol)	(mmol)	(mol L ⁻¹)
1.	carbon-blank	0	0	6
2.	PNA-0.05	0	0.05	6
3.	PNA-0.10	0	0.10	6
4.	PNA-0.15	0	0.15	6
5.	DMG-0.05-PNA-0.05	0.05	0.05	6
6.	DMG-0.05-PNA-0.10	0.05	0.10	6
7.	DMG-0.05-PNA-0.15	0.05	0.15	6
8.	DMG-0.10-PNA-0.05	0.10	0.05	6
9.	DMG-0.10-PNA-0.10	0.10	0.10	6
10.	DMG-0.10-PNA-0.15	0.10	0.15	6
11.	DMG-0.15-PNA-0.05	0.15	0.05	6
12.	DMG-0.15-PNA-0.10	0.15	0.10	6
13.	DMG-0.15-PNA-0.15	0.15	0.15	6

Notes:

- 1) In the case of DMG, it has been incorporated into carbon matrix instead of KOH electrolyte;
- 2) In the case of PNA, it has been just introduced KOH electrolyte;
- 3) All experiments were carried out in 100 mL 6 mol L⁻¹ KOH electrolyte.

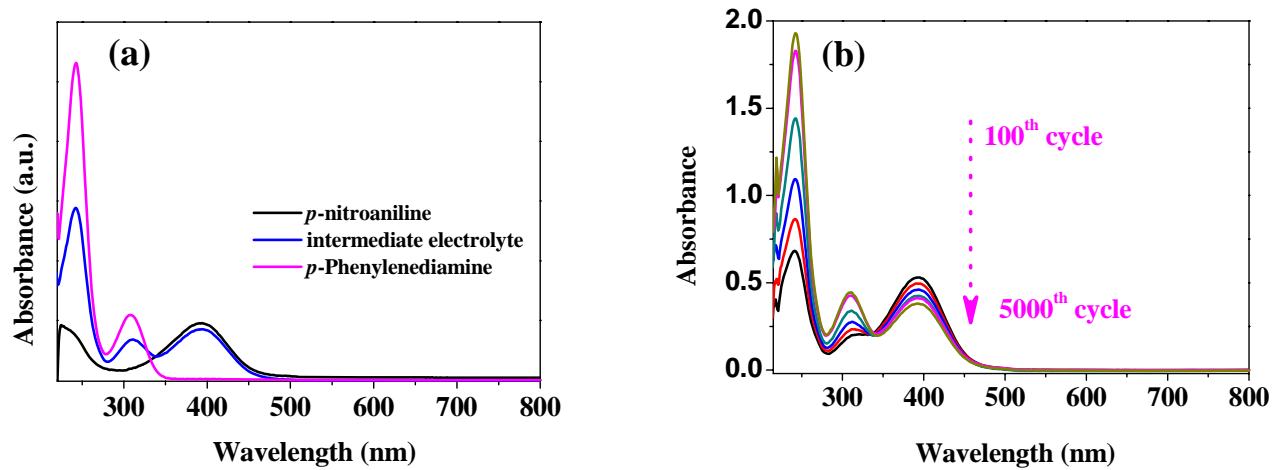


Fig. S1. (a) UV-vis absorption spectra of the *p*-nitroaniline, intermediate electrolyte, and *p*-phenylenediamine; (b) UV-vis absorption spectra of the mixed electrolyte in different stages of reaction.

Table S2. Observed decreases in capacitance for various samples after increasing the current density from 2 to 40 A g⁻¹.

NO.	Samples	Initial capacitance	Ultimate capacitance	Capacitance retention
		(F g ⁻¹) ^a	(F g ⁻¹) ^b	(%)
1.	carbon-blank	146.8	52.0	35.4
2.	PNA-0.05	180.8	84.0	46.5
3.	PNA-0.10	298.6	196.0	65.7
4.	PNA-0.15	226.2	128.0	56.6

Notes:

a: Specific capacitances calculated from GCD curves at a current density of 2 A g⁻¹.

b: Specific capacitances calculated from GCD curves at a current density of 40 A g⁻¹.

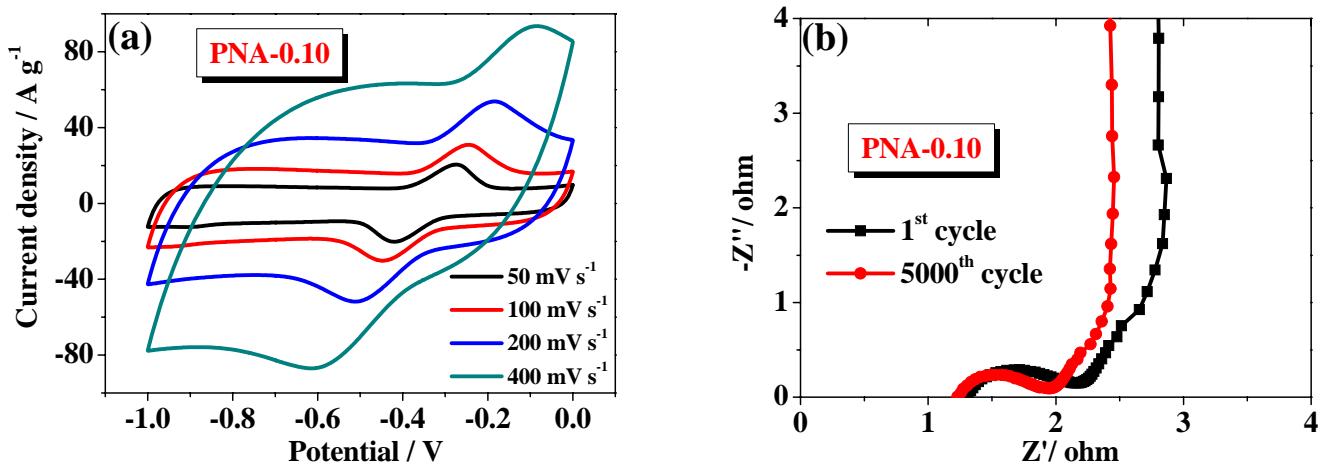


Fig. S2. The **PNA-0.10** sample measured in mixed electrolyte (6 mol L^{-1} KOH and 0.10 mmol PNA): (a) CV curves at different scan rates; (b) Nyquist plots before/after 5000 cycles.

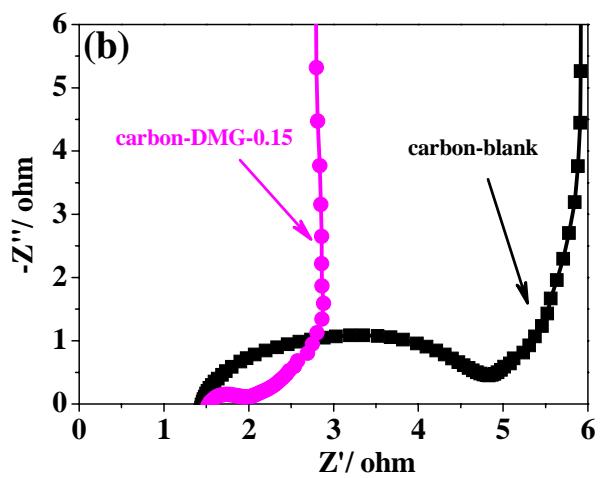
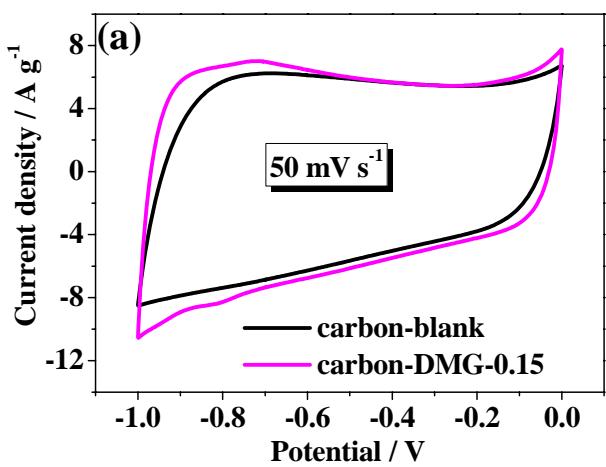


Fig. S3. The **carbon-blank** and **carbon-DMG-0.15** sample measured in a three-electrode system using 6 mol L^{-1} KOH: (a) CV curves at a scan rate of 50 mV s^{-1} ; (b) Nyquist plots.

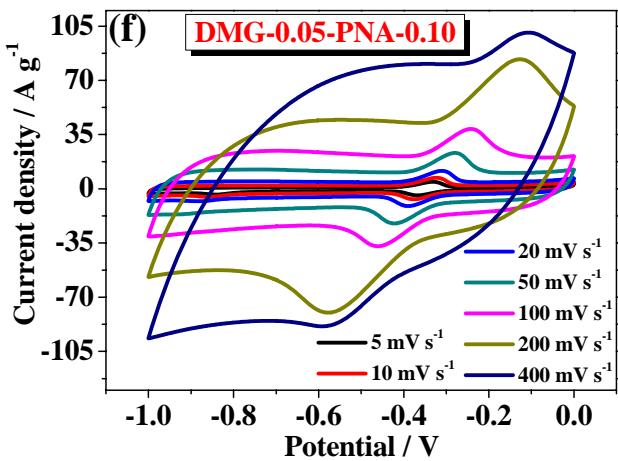
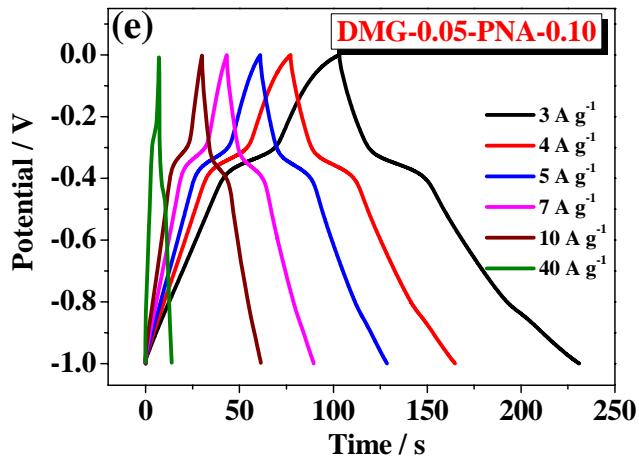
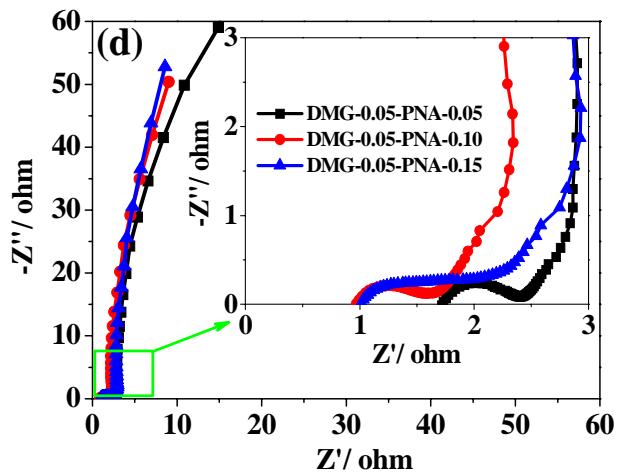
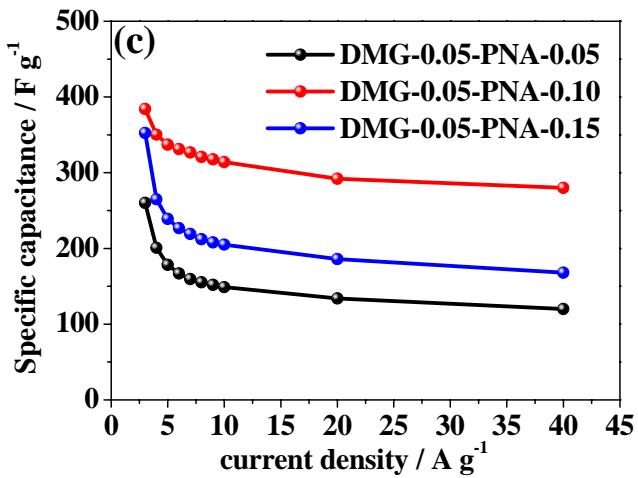
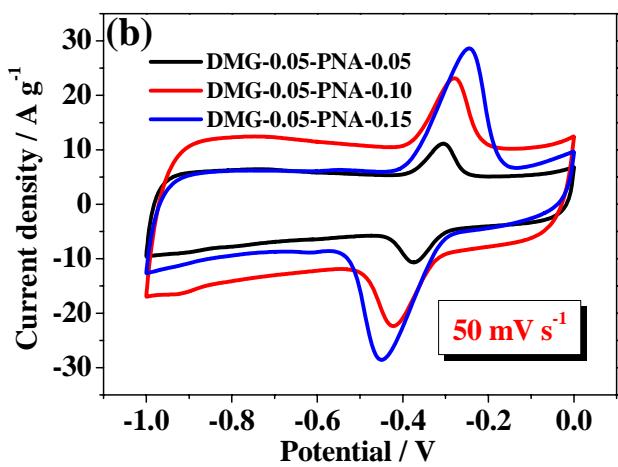
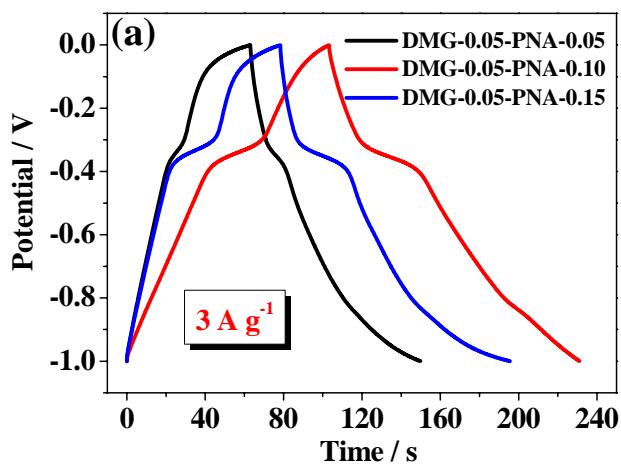


Fig. S4. The DMG-0.05-PNA-0.05/0.10/0.15 samples measured in a three-electrode

system using mixed electrolyte (6 mol L⁻¹ KOH and 0.05/0.10/0.15 mmol PNA): (a) GCD curves at a current density of 3 A g⁻¹; (b) CV curves at a scan rate of 50 mV s⁻¹; (c) specific capacitances calculated from GCD curves; (d) Nyquist plots as well as the magnified ones (the inset); (e) GCD curves of the **DMG-0.05-PNA-0.10** sample at different current densities; (f) CV curves of the **DMG-0.05-PNA-0.10** at different scan rates.

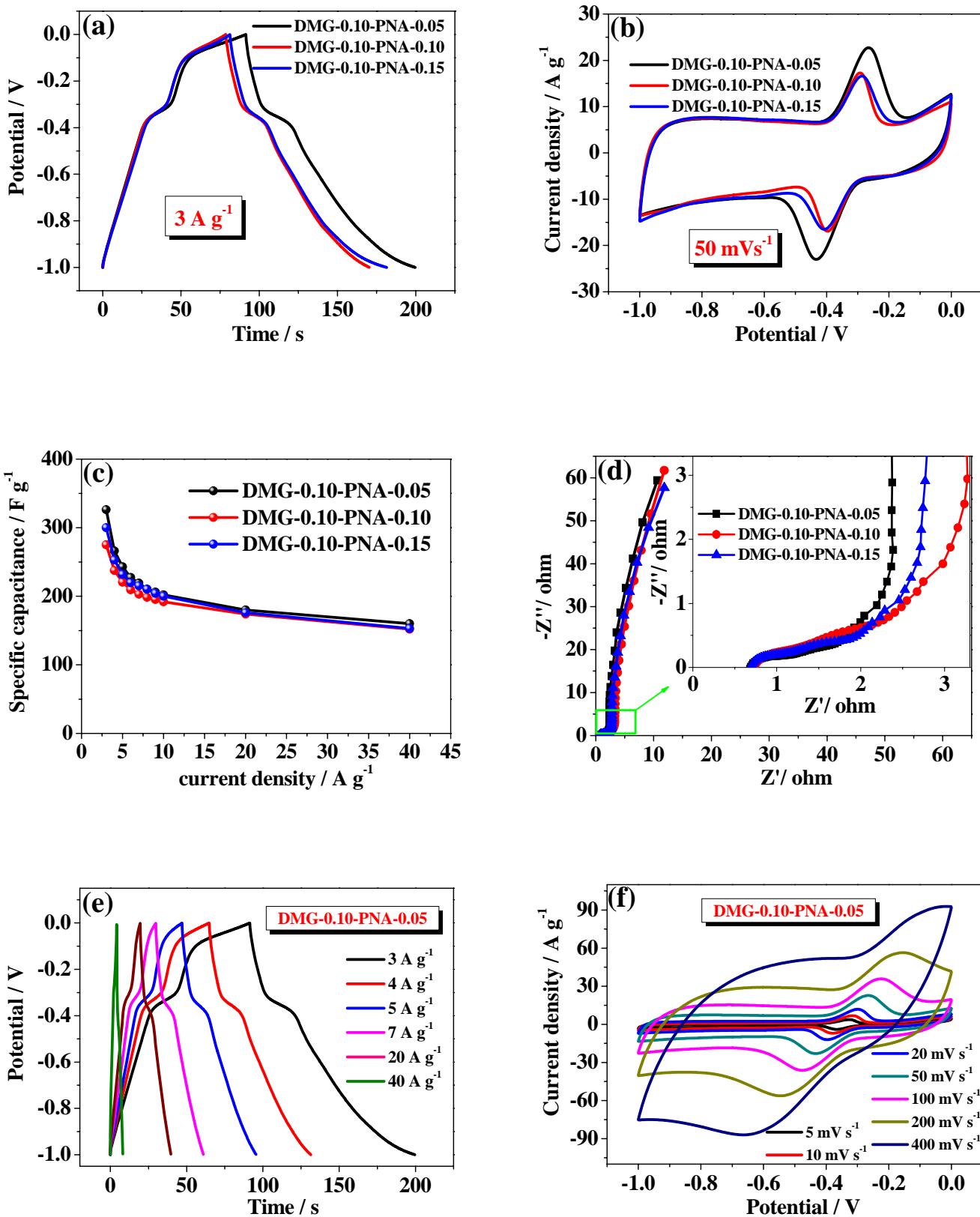


Fig. S5. The DMG-0.10-PNA-0.05/0.10/0.15 samples measured in a three-electrode

system using mixed electrolyte (6 mol L⁻¹ KOH and 0.05/0.10/0.15 mmol PNA): (a) GCD curves at a current density of 3 A g⁻¹; (b) CV curves at a scan rate of 50 mV s⁻¹; (c) specific capacitances calculated from GCD curves; (d) Nyquist plots; (e) GCD curves of the **DMG-0.10-PNA-0.05** sample at different current densities; (f) CV curves of the **DMG-0.10-PNA-0.05** at different scan rates.

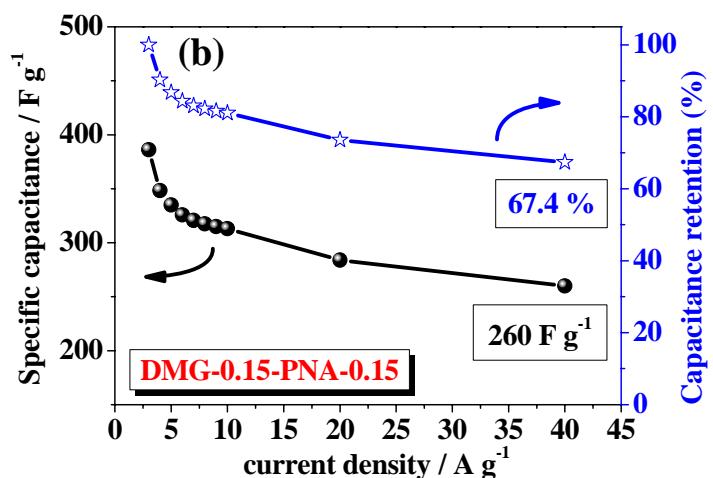
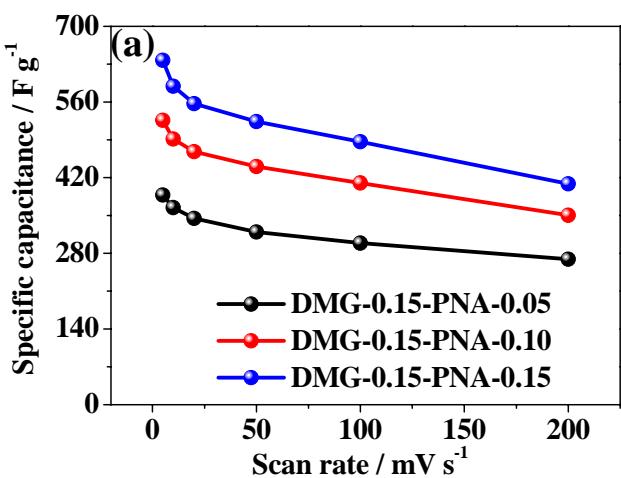


Fig. S6. The **DMG-0.15-PNA-0.05/0.10/0.15** samples measured in a three-electrode system using mixed electrolyte (6 mol L^{-1} KOH with different amount of PNA acting as redox additive): (a) the variation of specific capacitance as a function of scan rate; the **DMG-0.15-PNA-0.15** sample: (b) specific capacitances and capacitance retention.

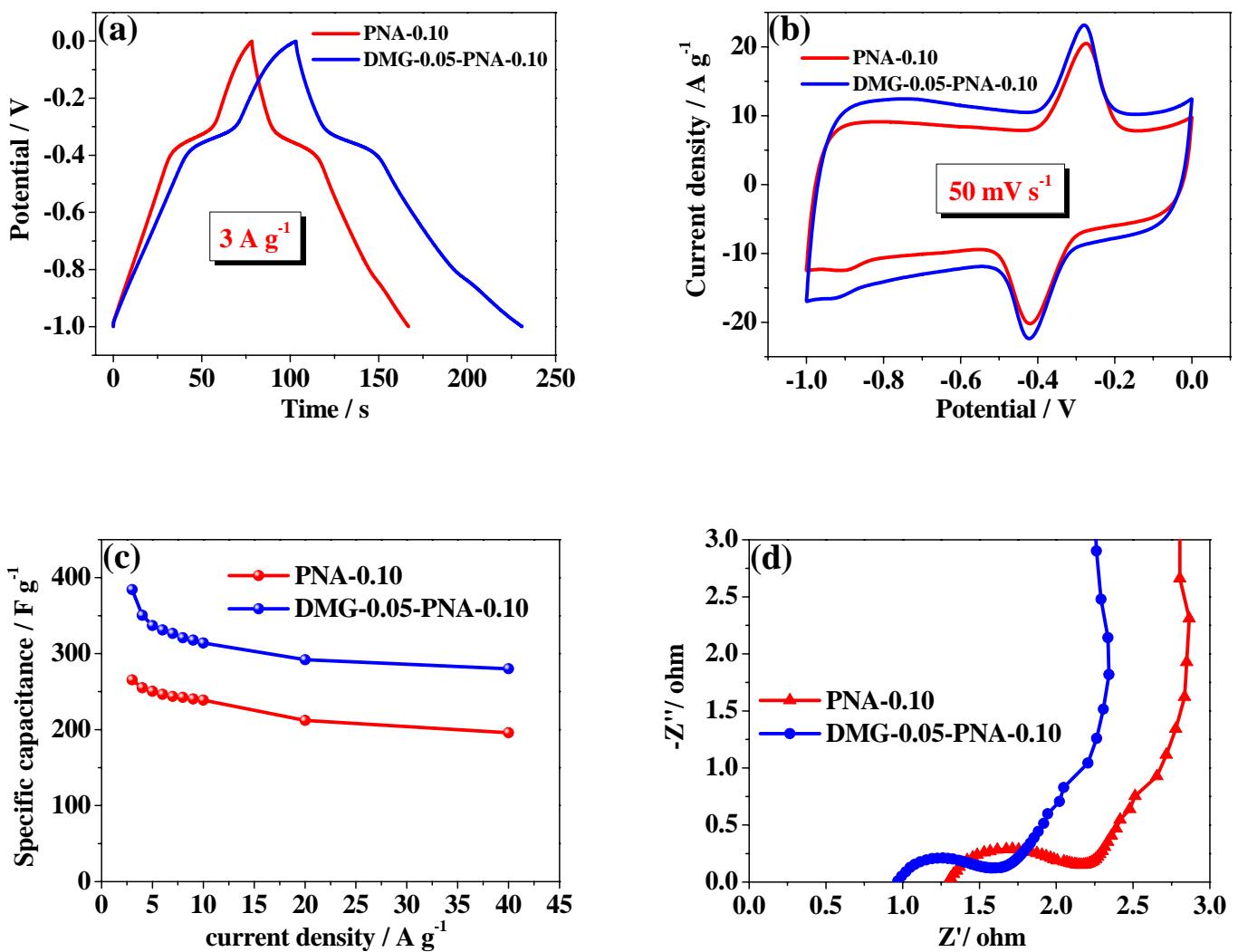


Fig. S7. The **PNA-0.10** and **DMG-0.05-PNA-0.10** samples measured in a three-electrode system: (a) GCD curves at a current density of 3 A g^{-1} ; (b) CV curves at a scan rate of 50 mV s^{-1} ; (c) specific capacitances calculated from GCD curves; (d) Nyquist plots as well as the magnified ones (the inset).

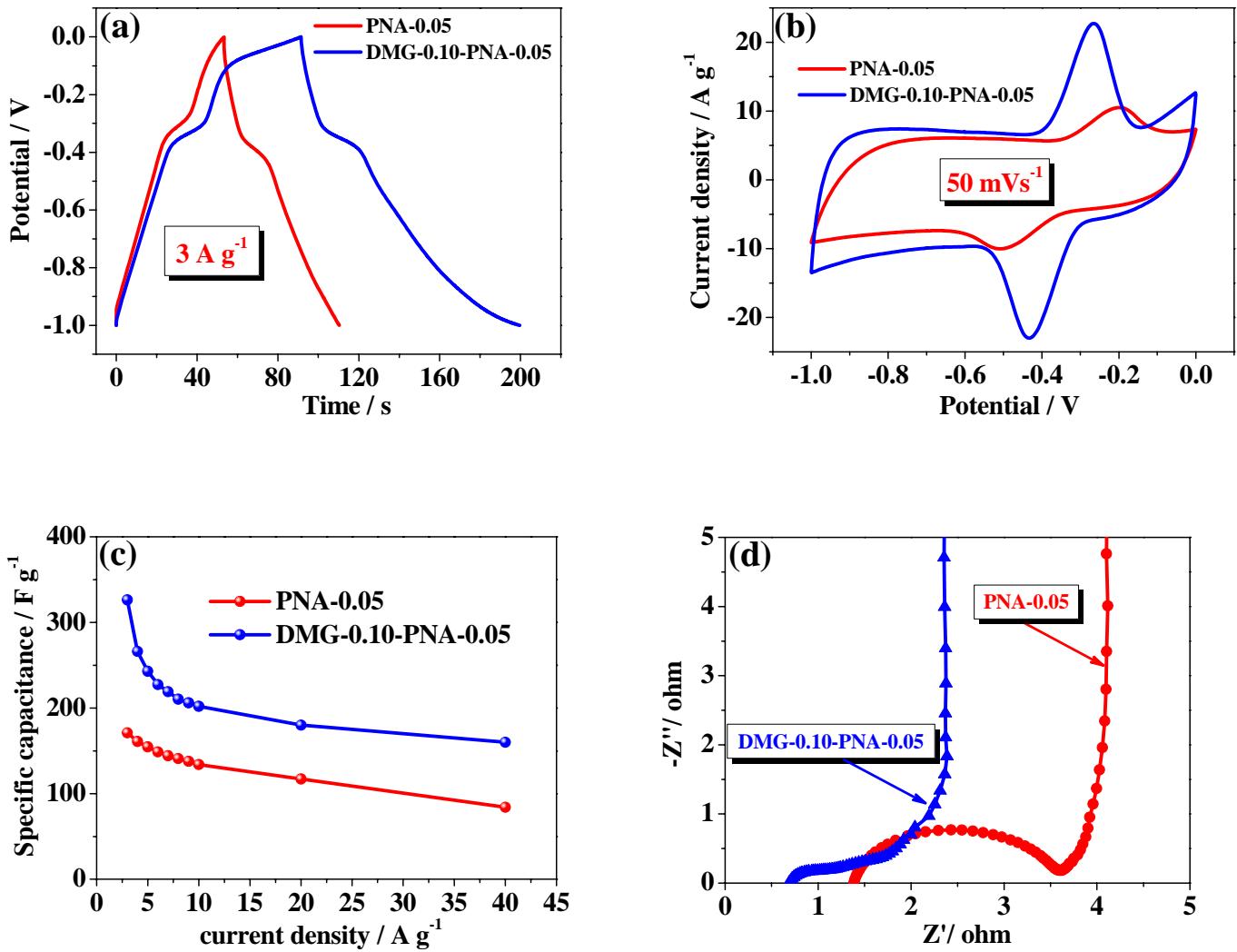


Fig. S8. The **PNA-0.05** and **DMG-0.10-PNA-0.05** samples measured in a three-electrode system: (a) GCD curves at a current density of 3 A g^{-1} ; (b) CV curves at a scan rate of 50 mV s^{-1} ; (c) specific capacitances calculated from GCD curves; (d) Nyquist plots.