

## Supporting Information

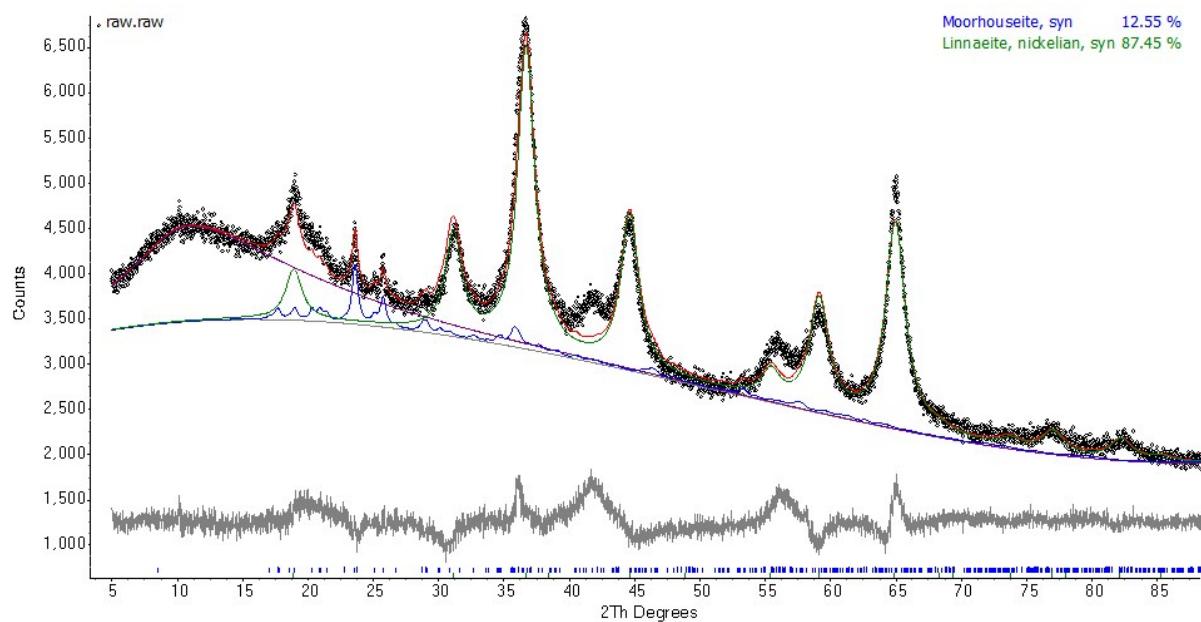
### **Plasma-induced on-surface sulfur vacancies in NiCo<sub>2</sub>S<sub>4</sub> enhance energy storage performance of supercapattories**

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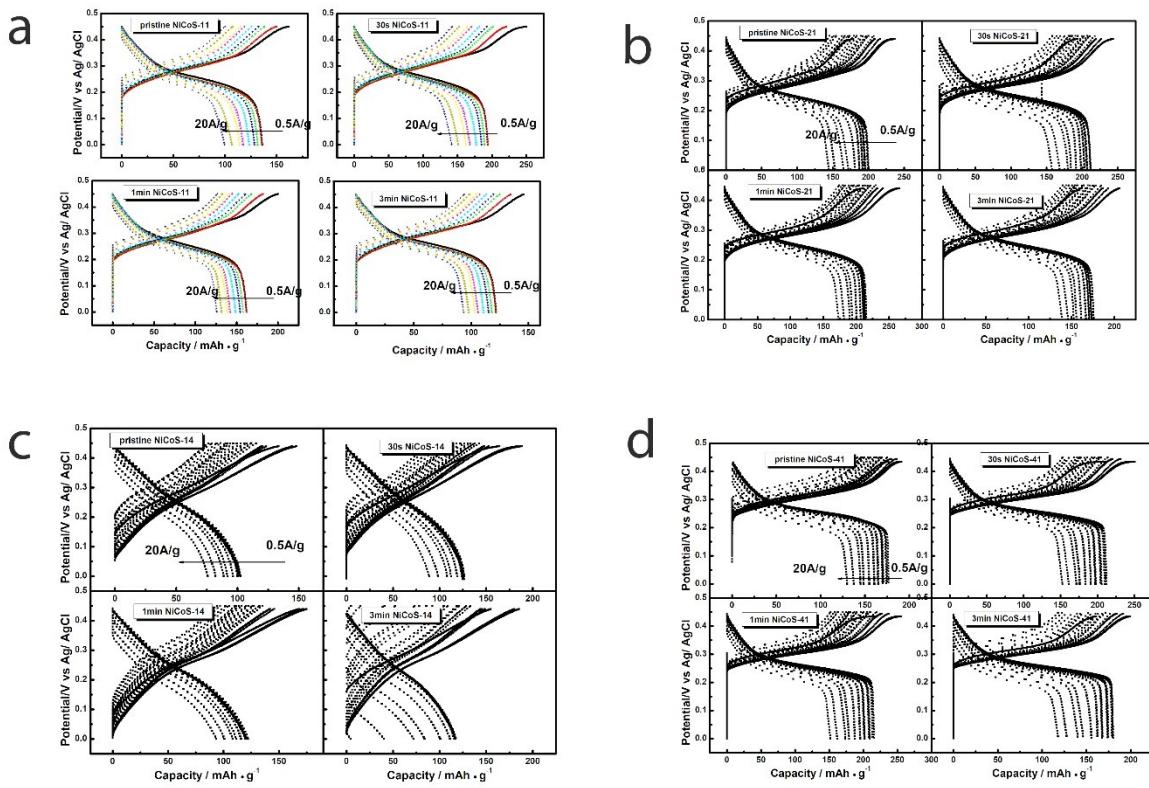
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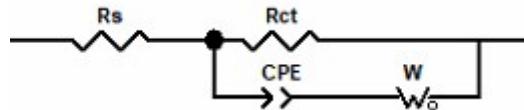
**Fig. S1** Fitted XRD pattern of pristine NiCoS-12

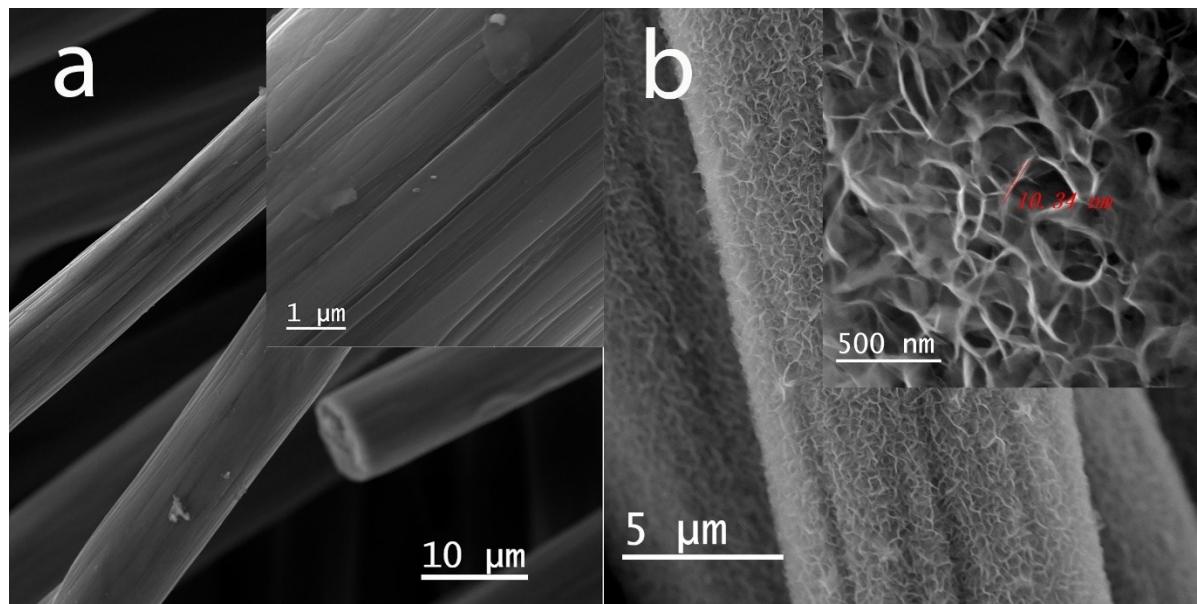


**Fig. S2** voltage vs. specific capacity of comparison group (various ratio of Ni/Co) with different plasma treatment duration **a**, NiCoS-11/CFC; **b**, NiCoS-21/CFC; **c**, NiCoS-14/CFC; **d**, NiCoS-41/CFC;

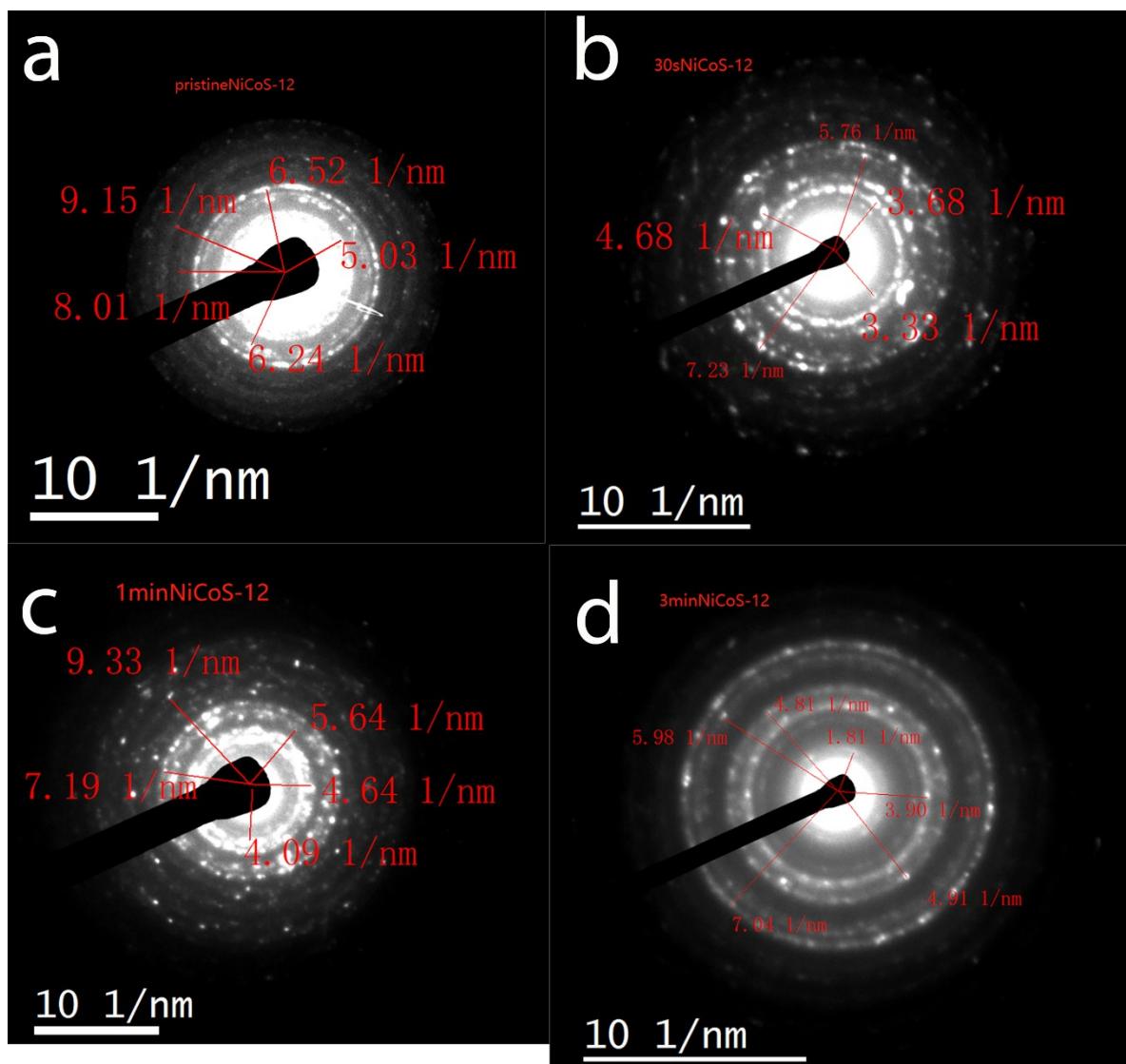
**Table S1.** EIS fitting results and equivalent circuit

sample	Rs ( $\Omega$ )	Rct ( $\Omega$ )
Pristine NiCoS-12	2.491	0.378
30s NiCoS-12	1.809	0.555
1min NiCoS-12	2.248	1.434
3min NiCoS-12	2.003	0.577





**Fig. S3** SEM images of **a**, bare CFC **b**, NiCoLDHs/CFC



**Fig. S4 a-d** SAED pattern of pristine NiCoS-12/CFC, 30s NiCoS-12/CFC, 1min NiCoS-12/CFC, 3min NiCoS-12/CFC

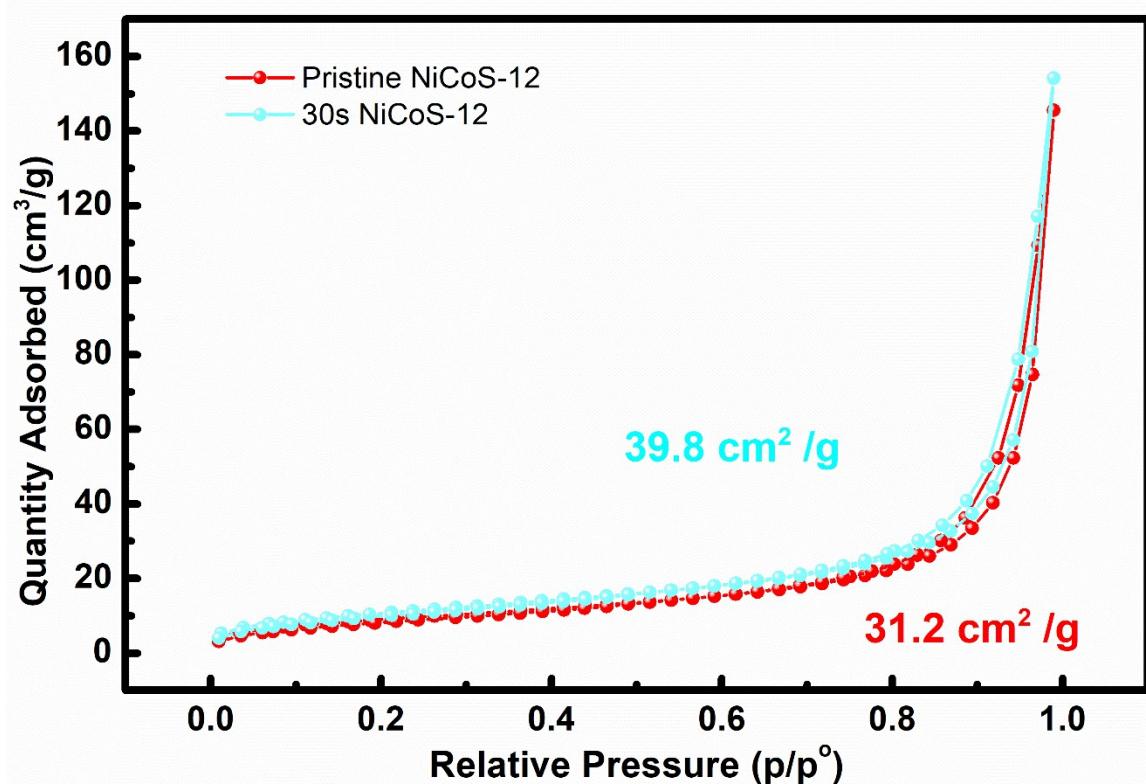


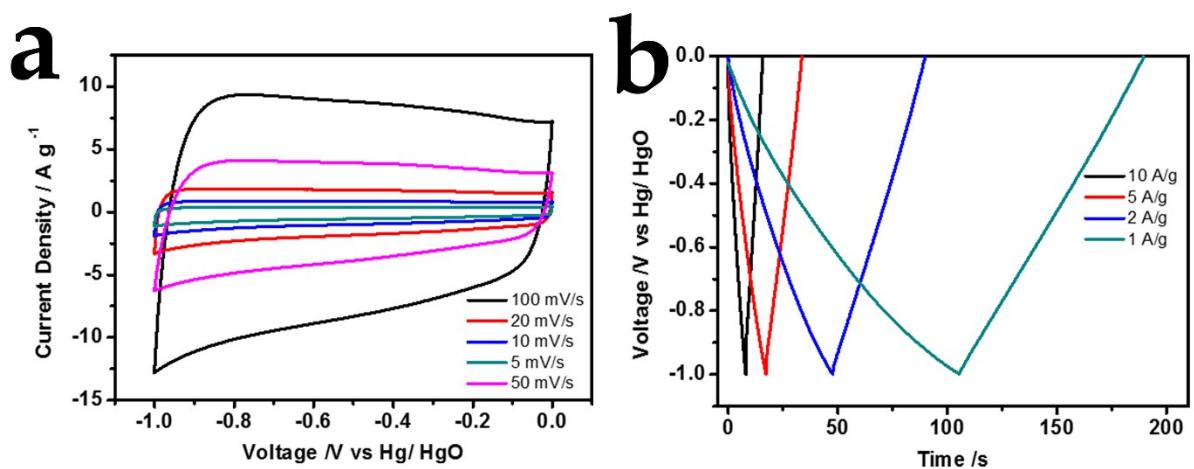
Fig. S5 N2 sorption isotherms of pristine and 30s NiCoS-12

**Table S2.** XPS survey spectrum of the NiCoS-12/ CFC with different duration plasma treatment and the elements atomic ratios.

XPS elements	Pristine	30s	1min	3min
Atomic ratio in sample	NiCoS-12	NiCoS-12	NiCoS-12	NiCoS-12
Ni (%)	2.79	2.85	2.87	1.63
Co (%)	5.86	5.64	5.04	3.28
S (%)	<b>38.92</b>	<b>27.12</b>	<b>25.59</b>	<b>19.59</b>
C (%)	27.24	39.07	37.69	55.06
O (%)	25.18	25.32	28.82	20.44

**Table S3.** Compositional ratio of Ni<sup>2+</sup> / Ni<sup>3+</sup> and Co<sup>2+</sup> / Co<sup>3+</sup> calculated from fitted Ni, Co HRXPS spectra

Fitted compositional areal ratio	Pristine NiCoS-12	30 s NiCoS-12	1 min NiCoS-12	3 min NiCoS-12
Ni <sup>2+</sup> / Ni <sup>3+</sup>	1.329	1.371	1.484	1.820
Co <sup>2+</sup> / Co <sup>3+</sup>	1.078	1.231	1.272	1.344



**Figure S 6.** **a**, CVs and **b**, GCD plots of negative material YP-50F commercial carbon.