

***Electronic supplementary information for***

**50–100- $\mu\text{m}$ -thick pseudocapacitive electrodes of  $\text{MnO}_2$  nanoparticles uniformly electrodeposited in carbon nanotube papers**

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**Table S1** Summary of structures and capacitances of all MnO<sub>2</sub>-CNT hybrid electrodes.

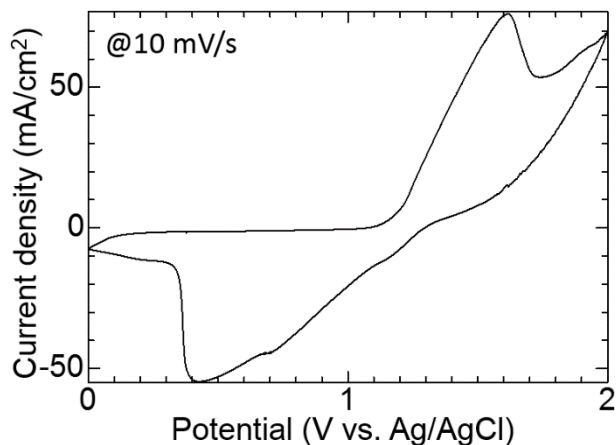
Condition	Before electrodeposition			After electrodeposition				Capacitance at 2–200 mV/s		
	Areal mass mg/cm <sup>2</sup>	Thickness μm	Density g/cm <sup>3</sup>	Areal mass mg/cm <sup>2</sup>	Thickness μm	Density g/cm <sup>3</sup>	MnO <sub>2</sub> wt%	Areal F/cm <sup>2</sup>	Total F/g	MnO <sub>2</sub> - based F/g
<b>Pristine</b>	1.00	25.0	0.40					0.02–0.01	24–13	
<b>CP 1.2 V</b>										
2 min	1.18	29.7	0.40	1.98	46.0	0.43	40.3	0.16–0.12	80–61	164–131
10 min	1.23	30.0	0.41	6.75	84.7	0.80	81.8	0.81–0.34	120–51	142–59
30 min	1.18	30.7	0.39	8.52	86.0	0.99	86.1	1.03–0.29	121–34	137–38
<b>CP 1.3 V</b>										
2 min	1.25	31.7	0.39	4.01	47.3	0.85	68.9	0.45–0.27	112–67	152–91
10 min	0.57	18.3	0.31	5.22	47.3	1.10	89.0	0.32–0.18	61–35	66–38
30 min	0.75	19.0	0.39	6.24	54.7	1.14	88.0	0.39–0.24	63–39	68–42
<b>CC 2 mA/cm<sup>2</sup></b>										
10 min	1.28	31.0	0.41	1.40	33.0	0.42	8.5	0.07–0.04	51–26	346–161
30 min	1.43	30.0	0.48	1.92	38.0	0.51	25.5	0.11–0.08	56–40	153–121
90 min	1.14	28.0	0.41	4.64	63.0	0.74	75.4	0.37–0.22	81–48	99–59
180 min	1.13	29.0	0.39	8.52	60.0	1.42	86.7	0.52–0.25	61–29	66–31
<b>CC 20 mA/cm<sup>2</sup></b>										
9 min	1.21	30.0	0.40	6.26	77.0	0.81	80.6	0.72–0.33	114–53	135–63
18 min	0.97	33.7	0.29	7.54	108.0	0.70	87.1	0.87–0.22	116–29	130–32
<b>Pulse 2.0 V</b>										
×15	1.03	32.7	0.32	2.61	71.9	0.36	60.3	0.25–0.17	98–65	167–100
×30	0.85	31.8	0.27	3.88	74.7	0.52	78.0	0.35–0.22	90–55	109–67
×45	1.02	36.2	0.28	5.17	104.0	0.50	80.3	0.51–0.30	99–57	117–68
×60	0.96	37.1	0.26	6.59	110.0	0.60	85.4	0.62–0.29	94–44	106–49

Thickness was measured by micrometre and thus somewhat larger (due to the surface roughness of the samples) than the value by SEM in Table S2.

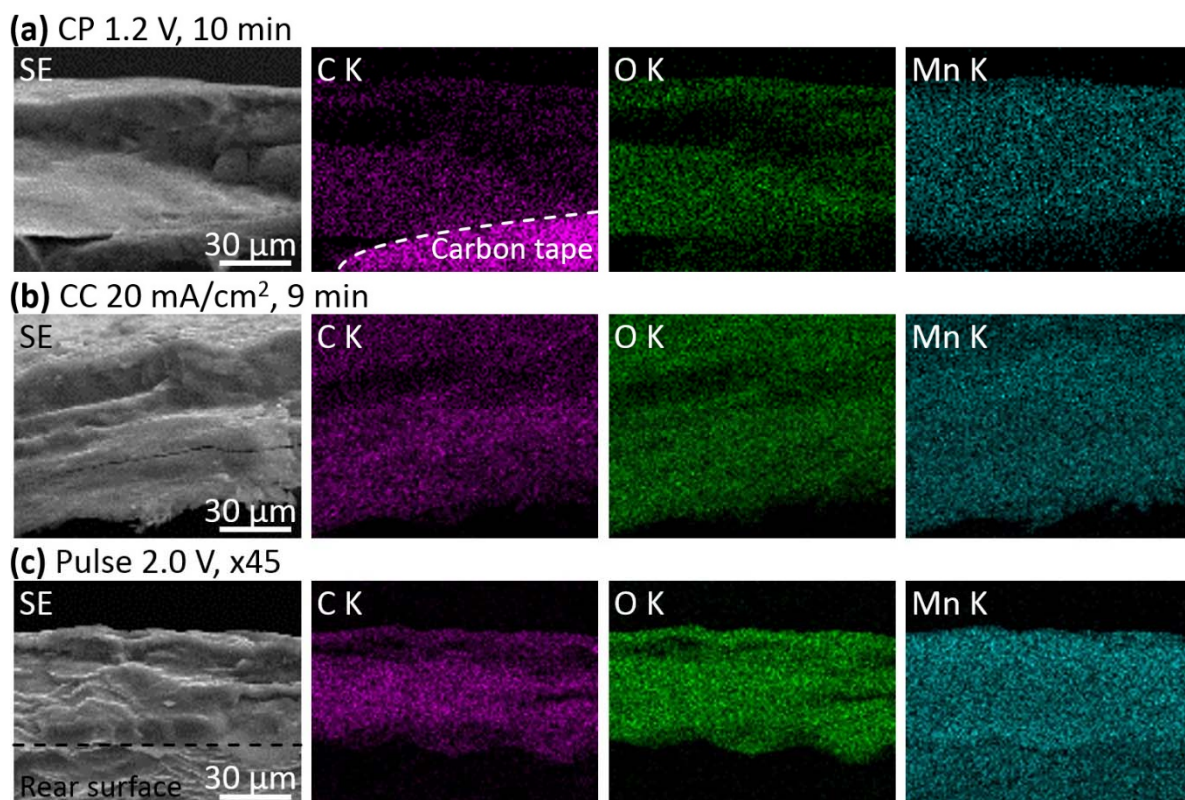
**Table S2** Structures and capacitances of the representative MnO<sub>2</sub>-CNT hybrid electrodes.

Condition	After electrodeposition				Capacitance at 2–200 mV/s		
	areal mass mg/cm <sup>2</sup>	thickness μm	density g/cm <sup>3</sup>	MnO <sub>2</sub> wt%	Areal F/cm <sup>2</sup>	Gravimetric F/g	Volumetric F/cm <sup>3</sup>
CP 1.2 V, 10 min	6.75	62.0	1.088	81.8	0.81–0.34	120–51	131–56
CC 20 mA/cm <sup>2</sup> , 9 min	6.26	71.5	0.876	80.6	0.72–0.33	114–53	100–46
Pulse x45	5.17	53.5	0.967	80.3	0.51–0.30	99–57	96–55

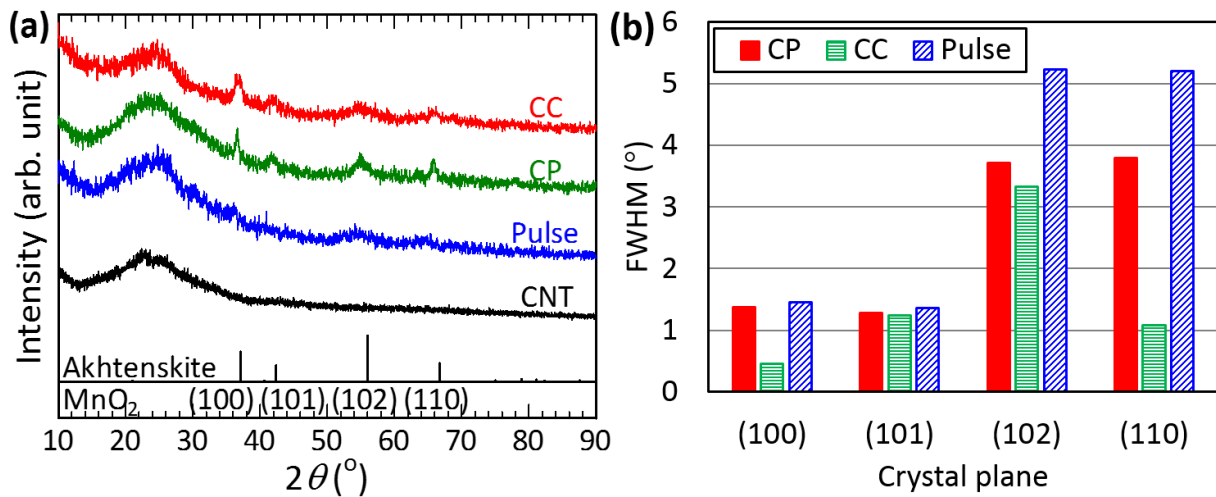
Thickness was measured by cross-sectional SEM and thus somewhat smaller than the value by micrometre in Table S1.



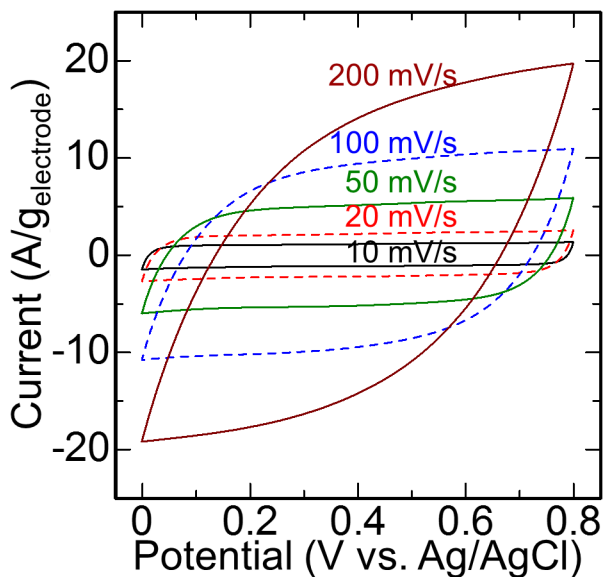
**Fig. S1** Typical current behavior during electrodeposition of MnO<sub>2</sub> in 0.6 M MnSO<sub>4</sub>/0.8 M H<sub>2</sub>SO<sub>4</sub> aqueous electrolyte at a scan rate of 10 mV/s.



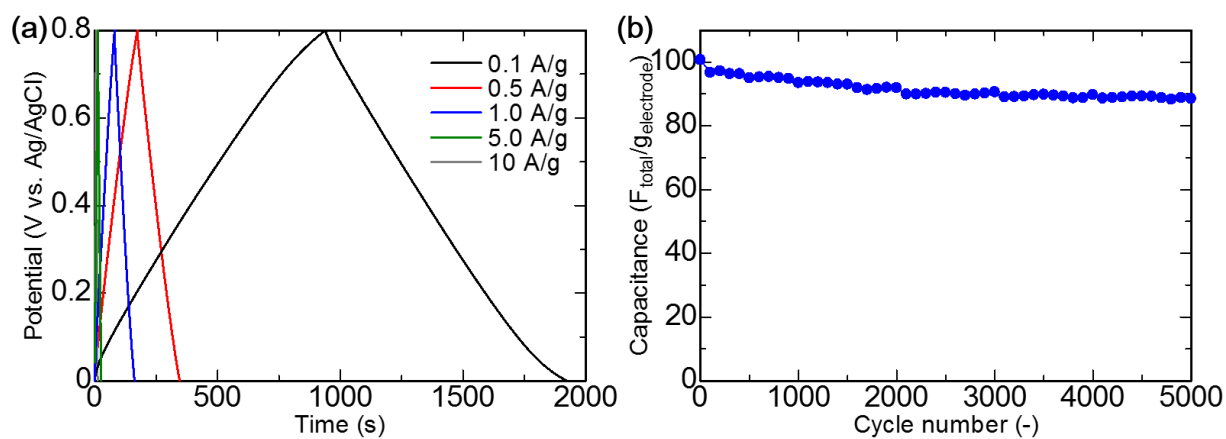
**Fig. S2** SEM-EDS elemental mapping of the cross-section of the representative MnO<sub>2</sub>-CNT hybrids taken at an acceleration voltage of 18 kV and magnification of x1,000. It is clearly shown that Mn is uniformly deposited throughout the CNT paper.



**Fig. S3** (a) XRD spectra of CNT papers without and with MnO<sub>2</sub> nanoparticles and (b) FWHM values of the peaks of Akhtenskite MnO<sub>2</sub>.



**Fig. S4** Typical CV curves of the MnO<sub>2</sub>-CNT electrode measured in 1 M Na<sub>2</sub>SO<sub>4</sub> aqueous solution with a AC-CNT hybrid electrode as the counter electrode and an Ag/AgCl electrode as the reference. MnO<sub>2</sub> was electrodeposited by CP at 1.20 V for 10 min.



**Fig. S5** Galvanostatic charge–discharge test of a MnO<sub>2</sub>–CNT hybrid electrode deposited on 30- $\mu$ m-thick CNT paper by CP at 1.20 V for 10 min. (a) Time profiles of potential of the working MnO<sub>2</sub>–CNT electrode for different charge–discharge rates. (b) Cycle performance of the MnO<sub>2</sub>–CNT electrode measured with a charge–discharge rate of 1 A/g and potential range of 0.0–0.8 V.