# Supporting Information

## A durable high-energy implantable energy storage system with binder-free electrodes usable in body fluids

Ji Su Chae,[a] Hoomin Lee,[b] Sung-Hyun Kim,[c] Nilesh R. Chodankar,[d] Sung-Min Kang,[e] Seonghan Lee,[c] Jeong Han Lee,[a,f] Young-Kyu Han,[d] Wan-Seob Cho,*[c], Yun Suk Huh*[b] and Kwang Chul Roh*[a]

[a]Energy & Environmental Division, Korea Institute of Ceramic Engineering & Technology, Jinju-si, 52851, Republic of Korea

[b]Department of Biological Engineering, Biohybrid Systems Research Center (BSRC), Inha University, Incheon 22212, Republic of Korea

[c]Lab of toxicology, Department of Health Sciences, The Graduate School of Dong-A University, Busan, 49315, Republic of Korea

[d]Department of Energy and Material Engineering, Dongguk University-Seoul, Seoul 04620, South Korea

[e]Department of Green Chemical Engineering, Sangmyung University, Cheonan, Chungnam 31066, Republic of Korea

[f]Department of Materials Science and Engineering, Korea University, Seoul 02841, Republic of Korea

**Methods**

The electrochemical parameters for the prepared electrodes and developed supercapacitor were calculated using the following equations:

***Specific capacitance from GCD***

$$Specific\ capacitance\ (F/g) = \frac{I\int Vdt}{mV^2} \quad (1)$$

***Specific energy and specific power from GCD***

$$Specific\ energy\ (Wh/kg) = \frac{C.\Delta V^2}{2 \times 3.6} \quad (2)$$

$$Specific\ power\ (W/kg) = \frac{3600 \times specifc\ energy}{\Delta t} \quad (3)$$

***Energy efficiency form the GCD***

$$Energy\ efficiency\ (\%) = \frac{dicharge\ specific\ energy\ \times 100}{charge\ specific\ energy} \quad (4)$$

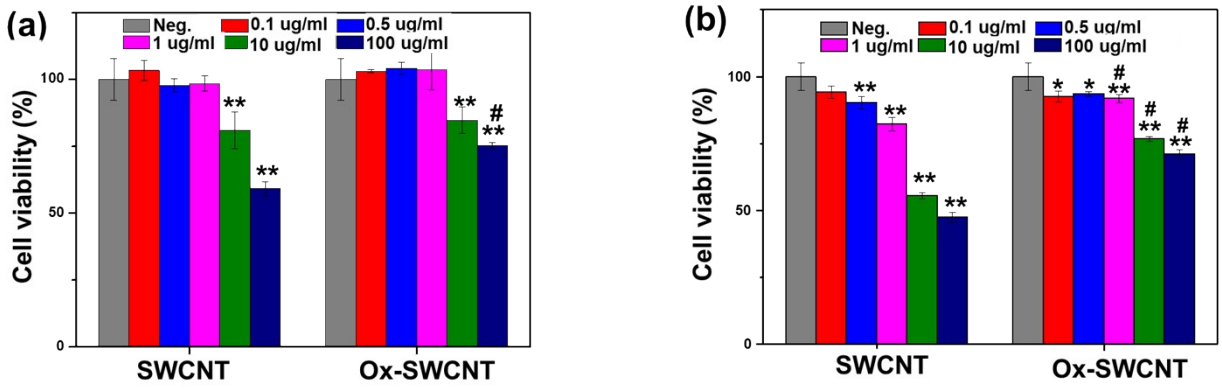


**Figure S1.** Cytotoxicity of dispersions of SWCNTs and Ox-SWCNTs for (a) COS-7 and (b) fibroblast cell lines measured using MTT assays. $^{*}p < 0.01$, $^{**}p < 0.001$ compared with the negative control. $^{\#}p < 0.01$ compared with SWCNTs at the same dose level.

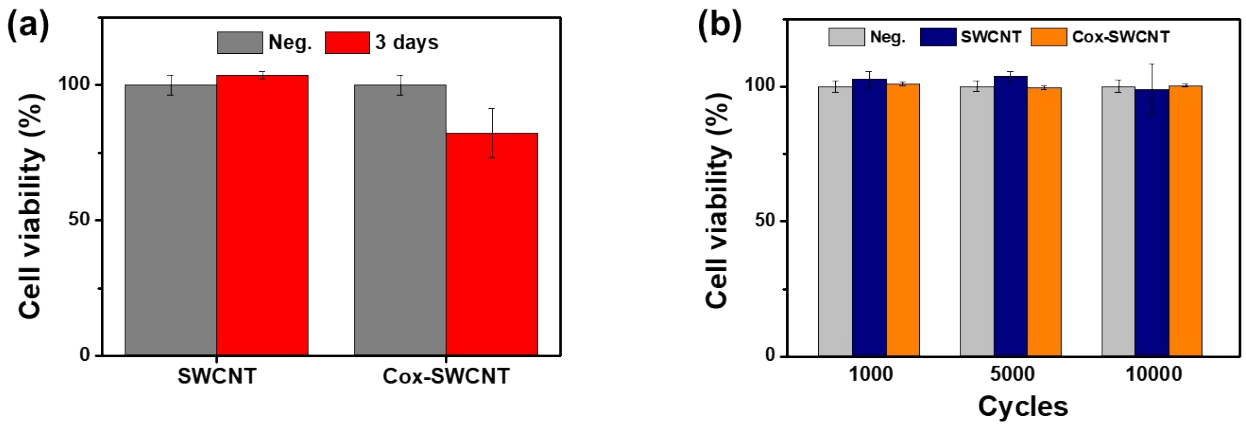


**Figure S2.** (a) Cell viability of human fibroblast cells measured using MTT assays after 3 days of exposure to SWCNT and Cox-SWCNT buckypaper electrodes. (b) Cell viability of human fibroblast cells measured using MTT assays after cyclic voltammetry from 1,000 to 10,000 cycles.

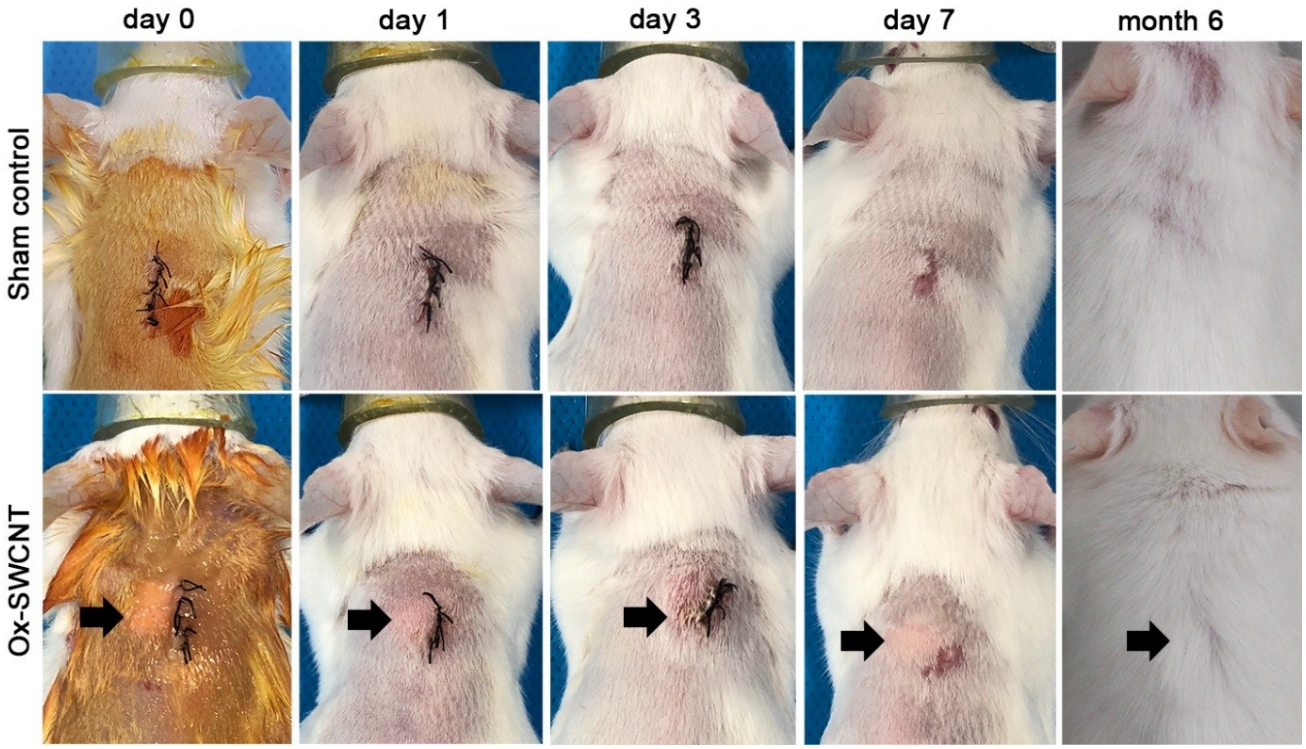


**Figure S3.** Representative gross lesions in mice after surgical implantation of Ox-SWCNTs. Ox-SWCNT buckypaper was satisfactorily implanted in the subcutaneous area (arrow) and produced no inflammatory reaction.

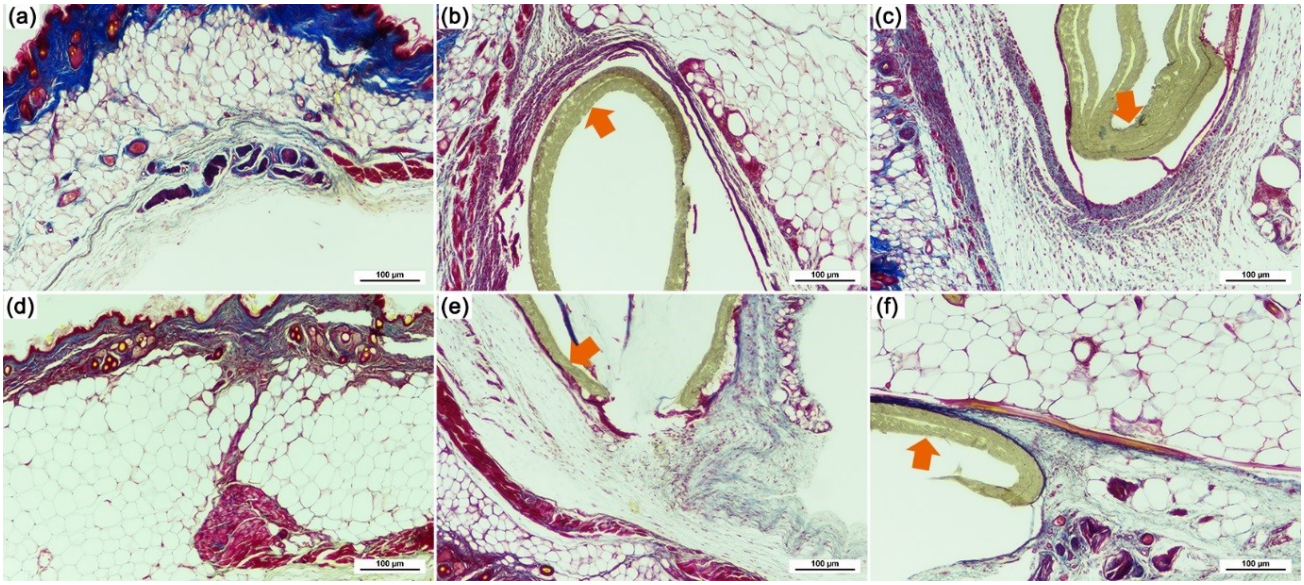


**Figure S4.** Representative images of Masson's trichrome staining at 7 days and 6 months after subcutaneous implantation of SWCNTs or Ox-SWCNTs. At 7 days after implantation, the (b) SWCNT and (c) Ox-SWCNT groups showed minimal-to-mild increases in collagen deposition (blue staining) near the Teflon tape (arrow) wrapping the electrodes, which contrasts the (a) sham control group. At 6 months after implantation, collagen deposition in the (e) SWCNT and (f) Ox-SWCNT groups was similar to that in the (d) sham control group. Scale bars = 100 µm.

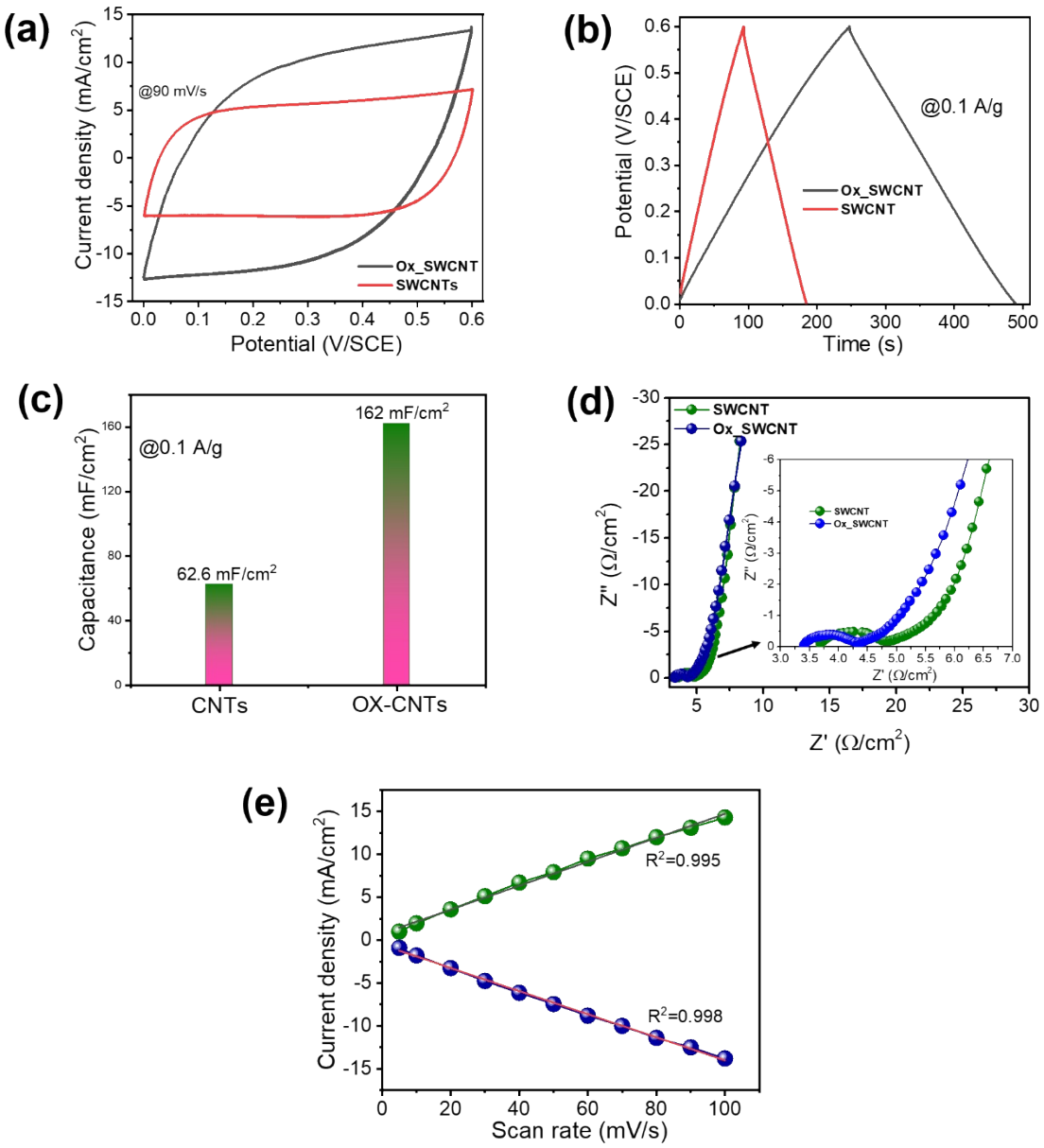


**Figure S5.** Three-electrode cell data; (a) cyclic voltammetry curves of SWCNT and Ox-SWCNT electrodes at a scan rate = 90 mV·s$^{-1}$ tested in simulated body fluid. (b) Galvanostatic charge-discharge profiles and (c) capacitance of SWCNT and Ox-SWCNT at 0.1A/g. (d) Nyquist plot and (e) plot of cathodic and anodic current at various scan rates.

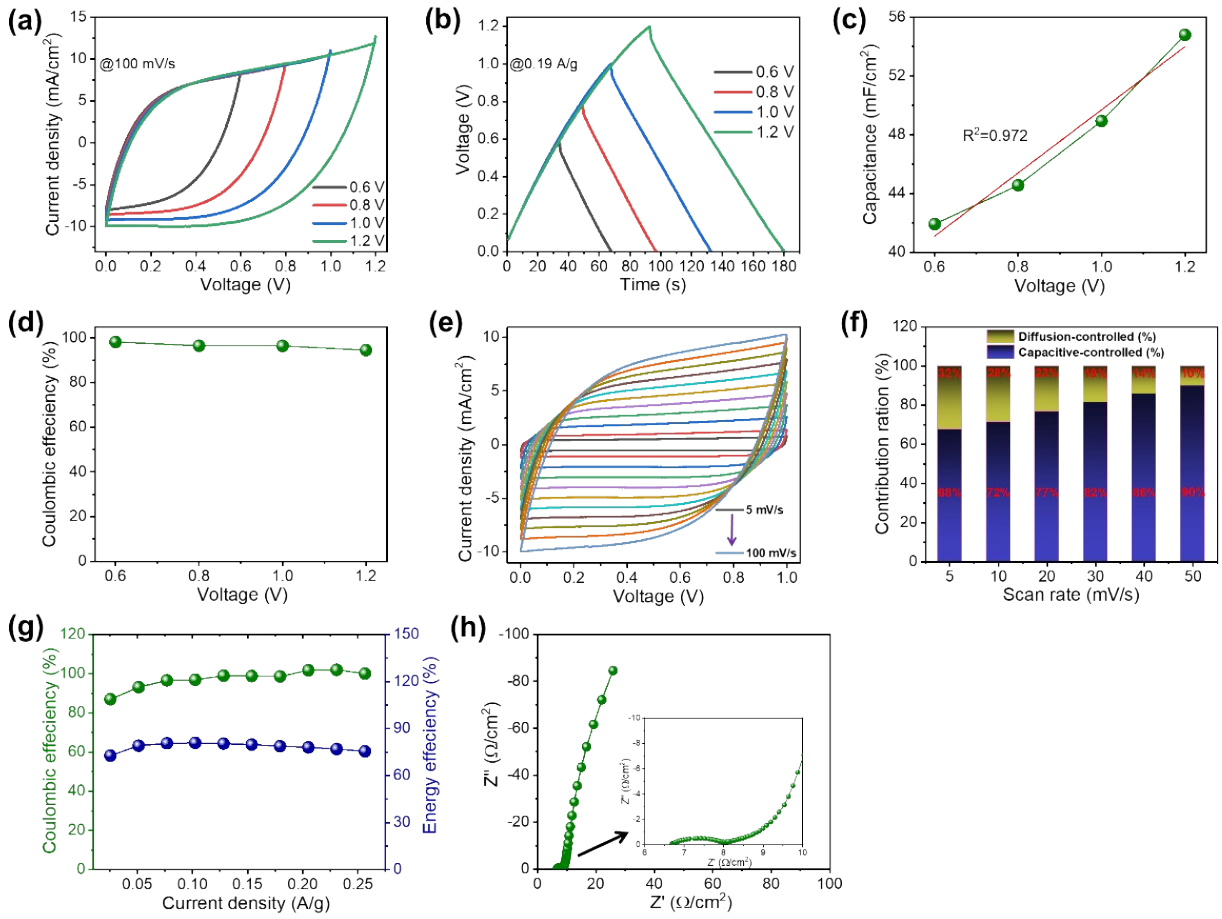


**Figure S6.** Two-electrodes of Ox-SWCNT cell data; (a) Cyclic voltammetry curves in different voltage windows at 100 mV/s can rate, (b) galvanostatic charge-discharge (GCD) curves in different voltage windows at 0.19 A/g current density, (c) plot of capacitance and (d) coulombic efficiency at different voltage windows derived from the GCD curves. (e) Cyclic voltammetry curves at various scan rates over 0–1.0 V. (f) Percentage capacitive contributions obtained at different scan rates of 5–50 mV $s^{-1}$, (g) coulombic and energy efficiency at various current density windows derived from the GCD curves, (h) Nyquist at identical conditions. Inset shows the magnified view of the Nyquist plot.

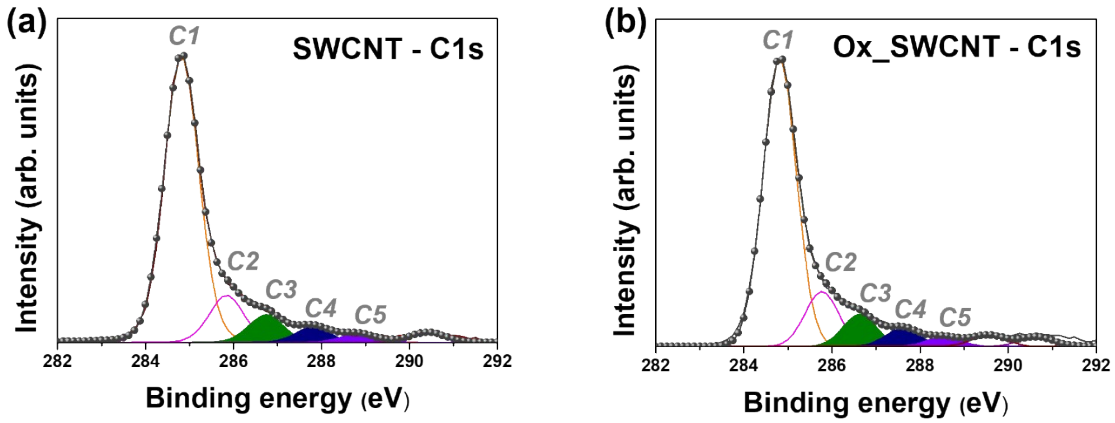


**Figure S7.** Deconvolution of high-resolution C1s X-ray photoelectron spectroscopy profiles of (a) SWCNT and (b) Ox-SWCNT buckypaper.

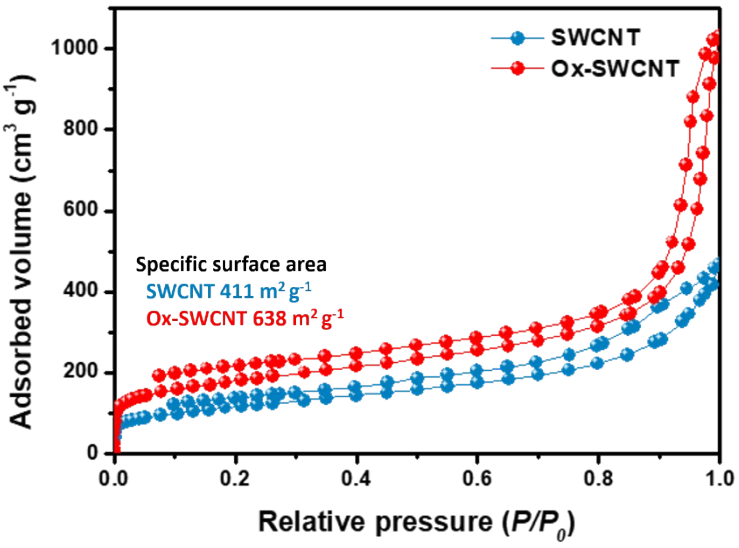


**Figure S8.** $N_2$ adsorption-desorption curves of SWCNT and Ox-SWCNT.

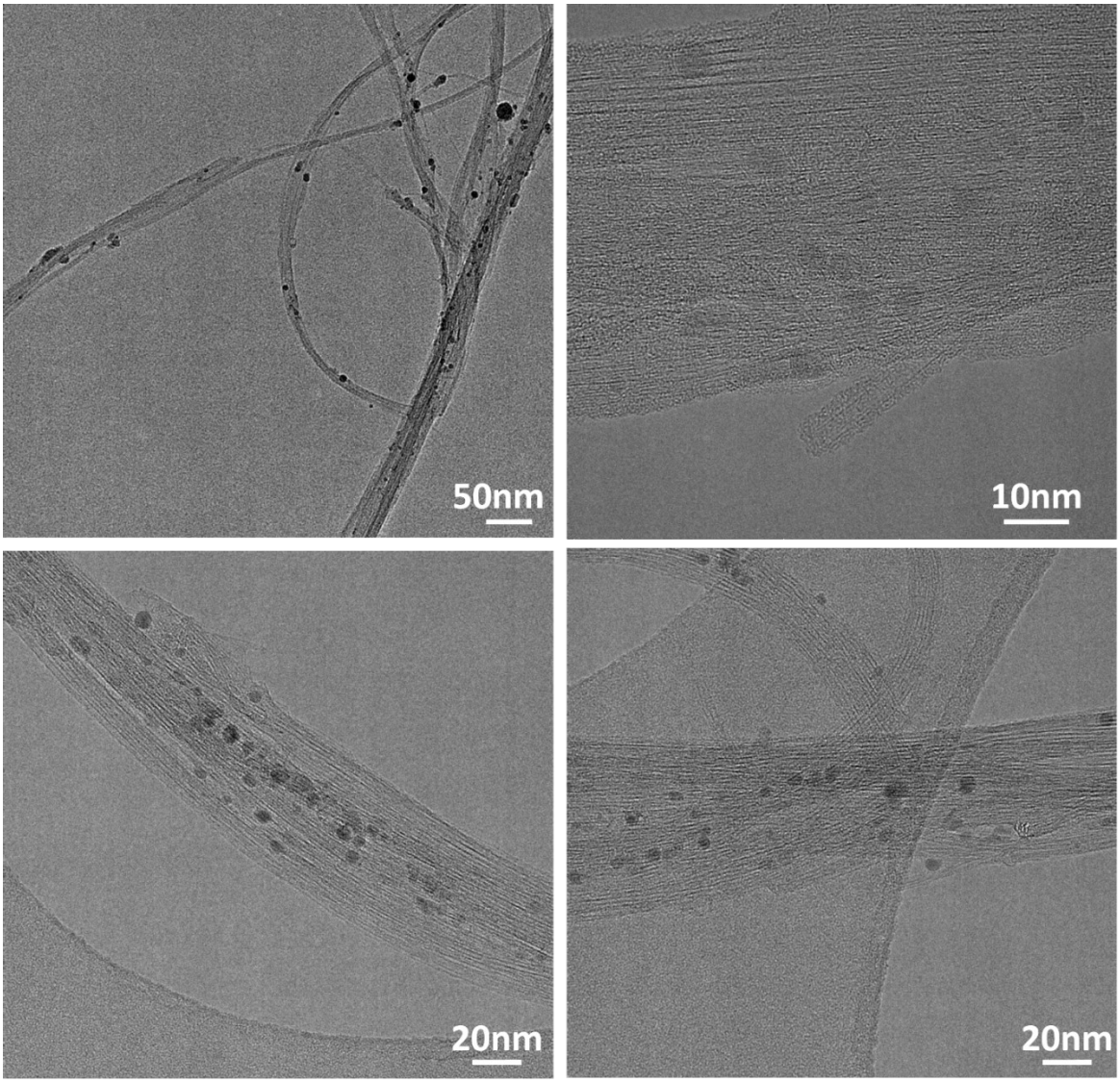


**Figure S9.** Cs-corrected transmission electron microscope (Cs-TEM)images of Ox-SWCNT for various point.

**Table S1.** Relative percentage of components on the sample surface

| Samples | Elemental composition (at. %) | | | | Element ratio | Concentration of chemical species (%) | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | (C1) | (C2) | (C3) | (C4) | (C5) | (C6) |
| | C | O | N | Fe | O/C | 284.8 eV –C=C | 285.7 eV –C–C | 286.6 eV –C–O | 287.6 eV –C=O | 288.5–289.5 eV –O–C=O | 291 eV π–π* |
| SWCNT | 96.87 | 1.63 | 0.66 | 0.84 | 0.02 | 72.61 | 11.94 | 7.03 | 3.87 | 2.11 | 2.44 |
| Ox-SWCNT | 93.17 | 5.96 | 0.58 | 0.28 | 0.06 | 68.00 | 12.99 | 7.61 | 4.3 | 4.87 | 2.23 |

**Table S2.** Hematological values of the mice at one week after implantation of the electrodes

| | Sham control | SWCNT | Ox-SWCNT |
|---|---|---|---|
| RBC (× $10^6$ cells/μL) | 8.35 ± 0.29[a)] | 8.48 ± 0.10 | 8.65 ± 0.31 |
| HGB (g/dL) | 13.56 ± 0.42 | 13.90 ± 0.54 | 13.94 ± 0.70 |
| HCT (%) | 47.74 ± 1.36 | 49.46 ± 1.54 | 48.00 ± 2.60 |
| MCV (fL) | 57.14 ± 1.36 | 58.32 ± 1.45 | 55.42 ± 1.81 |
| MCH (pg) | 16.22 ± 0.41 | 16.38 ± 0.53 | 16.08 ± 0.42 |
| MCHC (g/dL) | 28.38 ± 0.36 | 28.06 ± 0.54 | 29.02 ± 0.65 |
| RDW (%) | 14.22 ± 0.68 | 14.40 ± 0.60 | 13.94 ± 0.28 |
| MPV (fL) | 7.92 ± 0.44 | 7.42 ± 0.24 | 7.66 ± 0.39 |
| PLT (× $10^3$ cells/μL) | 1,050 ± 113.3 | 1,080 ± 112.4 | 1,077 ± 157.7 |
| WBC (× $10^3$ cells/μL) | 4.29 ± 0.41 | 7.51 ± 3.52 | 4.19 ± 0.73 |
| Neutrophils (%) | 15.86 ± 4.00 | 25.22 ± 13.91 | 21.10 ± 5.42 |
| Lymphocytes (%) | 72.80 ± 2.60 | 63.88 ± 14.40 | 71.90 ± 7.22 |
| Monocytes (%) | 1.70 ± 0.32 | 3.84 ± 1.40* | 2.12 ± 0.74 |
| Eosinophils (%) | 8.78 ± 3.53 | 5.78 ± 7.46 | 4.06 ± 1.69 |
| Basophils (%) | 0.30 ± 0.16 | 0.26 ± 0.09 | 0.10 ± 0.07 |

RBC: red blood cell count; HGB: hemoglobin; HCT: hematocrit; MCV: mean corpuscular volume; MCH: mean corpuscular hemoglobin; MCHC: mean corpuscular hemoglobin concentration; RDW: red cell distribution width; MPV: mean platelet volume; WBC: white blood cell count.

**a)** Data are expressed as mean ± SD (*$p < 0.01$).

**Table S3.** Hematological values of the mice at six months after implantation of the electrodes (related to **Fig. 5**)

| | Sham control | SWCNT | Ox-SWCNT |
|---|---|---|---|
| RBC (× $10^6$ cells/μL) | 9.78 ± 0.04[a)] | 9.32 ± 0.31 | 9.20 ± 0.52 |
| HGB (g/dL) | 15.70 ± 0.54 | 14.55 ± 0.27* | 14.85 ± 0.63 |
| HCT (%) | 53.10 ± 2.49 | 48.43 ± 1.11 | 49.05 ± 2.50 |
| MCV (fL) | 54.30 ± 0.70 | 52.03 ± 1.93 | 53.33 ± 0.71 |
| MCH (pg) | 16.10 ± 0.34 | 15.65 ± 0.63 | 16.18 ± 0.34 |
| MCHC (g/dL) | 29.63 ± 0.59 | 30.05 ± 0.42 | 30.33 ± 0.53 |
| RDW (%) | 14.00 ± 0.41 | 14.25 ± 0.25 | 14.45 ± 0.79 |
| MPV (fL) | 6.10 ± 0.22 | 6.30 ± 0.18 | 6.00 ± 0.33 |
| PLT (× $10^3$ cells/μL) | 1087 ± 43 | 1159 ± 171 | 1175 ± 110 |
| WBC (× $10^3$ cells/μL) | 4.73 ± 2.99 | 3.57 ± 1.64 | 3.24 ± 1.20 |
| Neutrophils (%) | 21.78 ± 3.98 | 19.20 ± 5.69 | 16.15 ± 4.47 |
| Lymphocytes (%) | 68.90 ± 5.42 | 74.30 ± 7.04 | 77.78 ± 6.53 |
| Monocytes (%) | 3.60 ± 0.94 | 2.00 ± 1.00 | 2.40 ± 0.62 |
| Eosinophils (%) | 5.00 ± 1.33 | 3.55 ± 1.41 | 2.88 ± 1.88 |
| Basophils (%) | 0.18 ± 0.10 | 0.20 ± 0.08 | 0.13 ± 0.10 |

RBC: red blood cell count; HGB: hemoglobin; HCT: hematocrit; MCV: mean corpuscular volume; MCH: mean corpuscular hemoglobin; MCHC: mean corpuscular hemoglobin concentration; RDW: red cell distribution width; MPV: mean platelet volume; WBC: white blood cell count.
[a)]Data are expressed as mean ± SD (*$p < 0.05$).

**Table S4.** Resistnace of SWCNT and Ox-SWCNT

| Electrode | Rs ($\Omega/cm^2$) | Rct ($\Omega/cm^2$) |
|---|---|---|
| SWCNT | 3.72 | 1.25 |
| Ox-SWCNT | 3.47 | 0.86 |

**Table S5**. Comparative electrochemical parameters for carbon non-bio SCs reported in the literature

| Electrode | Electrolyte | Voltage window (V) | Max. energy density | Max. power density | Cycling stability (%) | Ref. |
|---|---|---|---|---|---|---|
| Fe-ZnO | Hydrogel of PBS/PVA | 0–1 | 0.153 μWh/cm$^2$ | 0.526 mW/cm$^2$ | 70 over 3000 cycles | [1] |
| Oxidized Mo wire | NaCl/PVA gel | 0–0.8 | 0.370 μWh/cm$^2$ | 0.8 mW/cm$^2$ | 82 over 5000 cycles | [2] |
| Au/$MnO_2$/Au | PVA/$LiClO_4$ gel | 0–0.8 | 0.376 μWh/cm$^2$ | 0.099 mW/cm$^2$ | 81 over 1000 cycles | [3] |
| $MnO_2$@SiNWs | $LiClO_4$-PMPyrrBTA | 0–2.2 | 9.1 μWh/cm$^2$ | 0.388 mW/cm$^2$ | 91 over 5000 cycles | [4] |
| CNT/PANI | PMMA-PC-$LiClO_4$ gel | 0–0.8 | 0.004mWh/cm$^2$ | 0.07 mW/cm$^2$ | 87 over 20,000 cycles | [5] |
| $MnO_2$-CNT buckypaper | PVA/KOH gel | 0–1.0 | 4.2 μWh/cm$^2$ | 4 mW/cm$^2$ | 98 over 20,000 cycles | [6] |
| PEDOT:PSS/ ferritin/MWNT | PBS | 0–0.8 | 0.82 μWh/cm$^2$ | 150 μW/cm$^2$ | 98 over 1000 cycles. | [7] |
| Ox-SWCNT | SBF | 0–1 | 7.12 mWh/cm$^2$ (50 mW/cm$^2$) | 500 mW/cm$^2$ (6.52 mWh/cm$^2$) | 100 over 50,000 cycles | This work |

## References


1 H. Li, C. Zhao, X. Wang, J. Meng, Y. Zou, S. Noreen, L. Zhao, Z. Liu, H. Ouyang, P. Tan, *Adv. Sci.*, 2019, **6**, 1801625.

2 H. Lee, G. Lee, J. Yun, K. Keum, S. Y. Hong, C. Song, J. W. Kim, J. H. Lee, S. Y. Oh, D. S. Kim, *Chem. Eng. J.*, 2019, **366**, 62-71.

3 H. Hu, Z. Pei, H. Fan, C. Ye, *Small*, 2016, **12**, 3059-3069.

4 D. P. Dubal, D. Aradilla, G. Bidan, P. Gentile, T. J. Schubert, J. Wimberg, S. Sadki, P. Gomez-Romero, *Sci. Rep.*, 2015, **5**, 1-10.

5 L. Li, Z. Lou, W, Han, D. Chen, K. Jiang, G. Shen, *Adv. Mater. Technol.*, 2017, **2(3)**, 1600282.

6 L. Dong, C. Xu,Y. Li, Z. Pan,G. Liang, E. Zhou, F. Kang, Q.-H. Yang, *Adv. Mater.*, 2016, **28(42)**, 9313-9319.

7 H. J. Sim, C. Choi, D. Y. Lee, H. Kim, J. H. Yun, J. M. Kim, T. M. Kang, O. Raquel, R. H. Baughman, K. W. Chang, S. J. Kim, *Nano Energy*, 2018, **47**, 385-392.