SUPPORTING INFORMATION

Binder-Free Carbon Nanotube Electrode for Electrochemical Removal of Chromium

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Figure S1. Scanning electron micrographs of (a) a pristine carbon nanotube (CNT) electrode and the same electrode after being dropped face-down to ground from (b) 0.5 m, (c) 1 m, (d) 2 m, (e) 3 m, and (f) 4 m. CNTs in contact with ground are increasingly pressed as the dropping height and the intensity of impact increase. However, there is no discernible change of the structure of CNT arrays or evidence of broken CNTs. Scale bar: $100 \mu m$.



Figure S2. Optical micrographs of (a) a carbon nanotube (CNT) electrode immersed in a pH 7 solution for (b) 10 min and (c) 120 min, the same electrode after the solution pH has been adjusted from 7 to 4 for (d) 10 min and (e) 120 min, and the same electrode after the solution pH has been adjusted from 4 to 0 for (f) 10 min and (g) 120 min. The bright spots in the micrographs are light passing through the remaining openings that are not blocked by CNT arrays (cf. Figure 1c in the main text). Potential swelling or stripping of CNTs from the electrode would have resulted in the expansion of the openings, which is not observed in this series of micrographs. Instead, the total area of the bright spots accounts for a constant 3.3% of the total micrographic area throughout the experiments, confirming that there is no swelling or stripping of CNT arrays in solutions having pH from 0 to 7. Scale bar: 100 µm.



Figure S3. Specific surface area of carbon nanotube (CNT) arrays on CNT electrodes. (a) Isotherm of nitrogen (N₂) at 77 K. The isotherms are classified as type IV isotherms containing mesopores.¹⁻² The overlapping of the isotherms obtained with different CNTs suggest that specific surface area is independent of CNT length. (b) Pore size distribution calculated by the non-local density functional theory. The pore size distribution plot of the CNTs exhibits a wide pore size distribution ranging from 2 nm to 100 nm, which is common for CNT samples.³⁻⁴ The pore size below and above 10 nm can be attributed to the inner cavity of CNTs and the porous structure formed between CNTs, respectively.⁴⁻⁵ CNTs with different lengths are distinguished by colors: red, $5(\pm 1)$ µm; black, $24(\pm 4)$ µm.



Figure S4. Linear sweep voltammograms of CNT electrodes for water reduction. CNT electrodes with different CNT lengths are distinguished by colors: red, $5(\pm 1) \mu m$; blue, $14(\pm 1) \mu m$; pink, $24(\pm 4) \mu m$. Solution conditions: 10 g L⁻¹ Na₂SO₄ and pH 0. Scan rate: 10 mV s⁻¹. SHE: Standard hydrogen electrode.



Figure S5. Correlation of the specific electrochemical surface area of carbon nanotube (CNT) electrodes and their CNT mass fraction (fraction of CNT mass in the total mass of CNTs and SSM). The solid line represents a least-square regression, giving a slope of $0.85(\pm 0.28)$ with R² = 0.99.



Figure S6. Energy dispersive X-ray spectrum of the carbon-paper anode after being used in the electrochemical treatment of chromium. The absence of peaks around 5.5 keV confirms that there is little chromium, if any, adsorbed on the anode. Conditions for the electrochemical treatment: initial concentration, 9.8 mg L⁻¹; pH, 3; duration, 120 min; potential, -1.4 V; cathode CNT length, $L = 14(\pm 1) \mu m$.

References

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