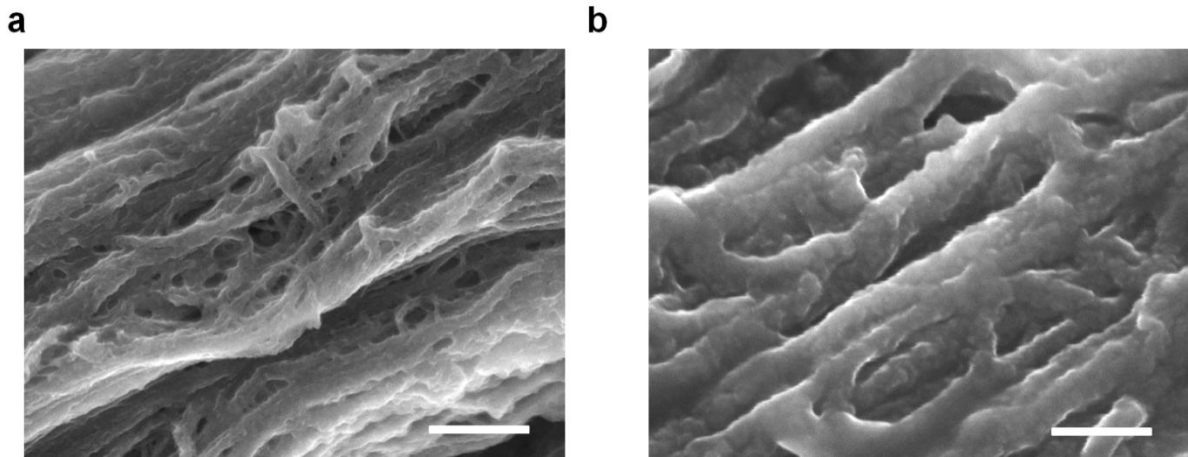
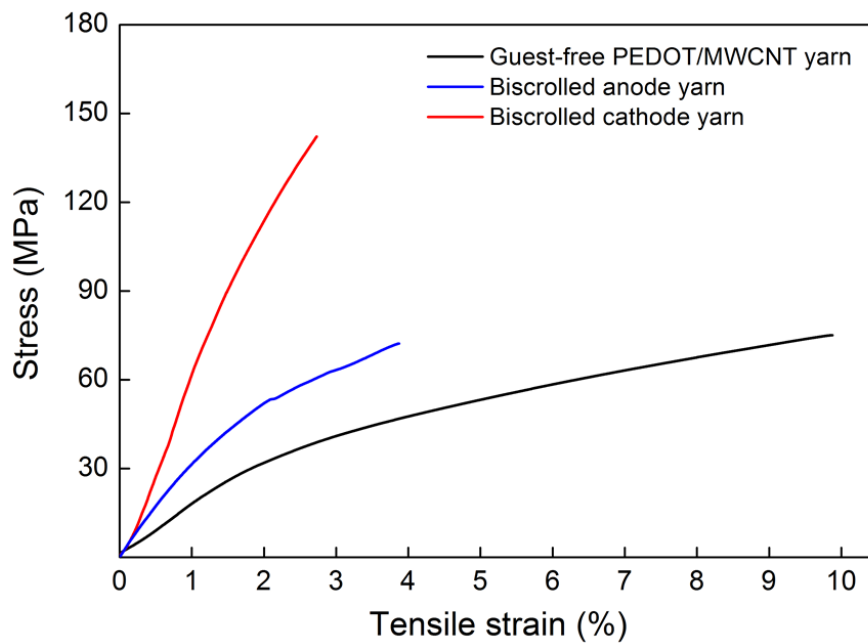


Supplementary Information

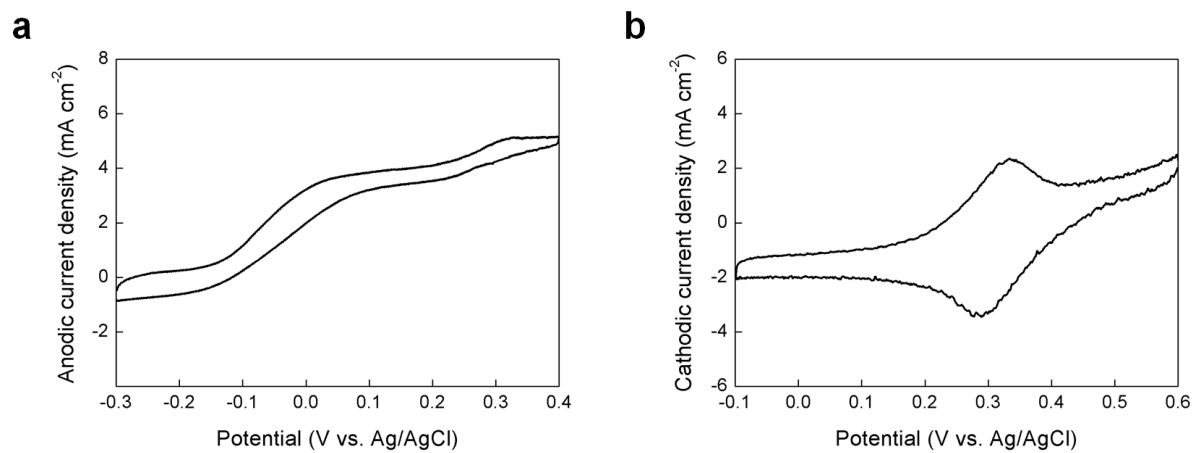
Supplementary Figures



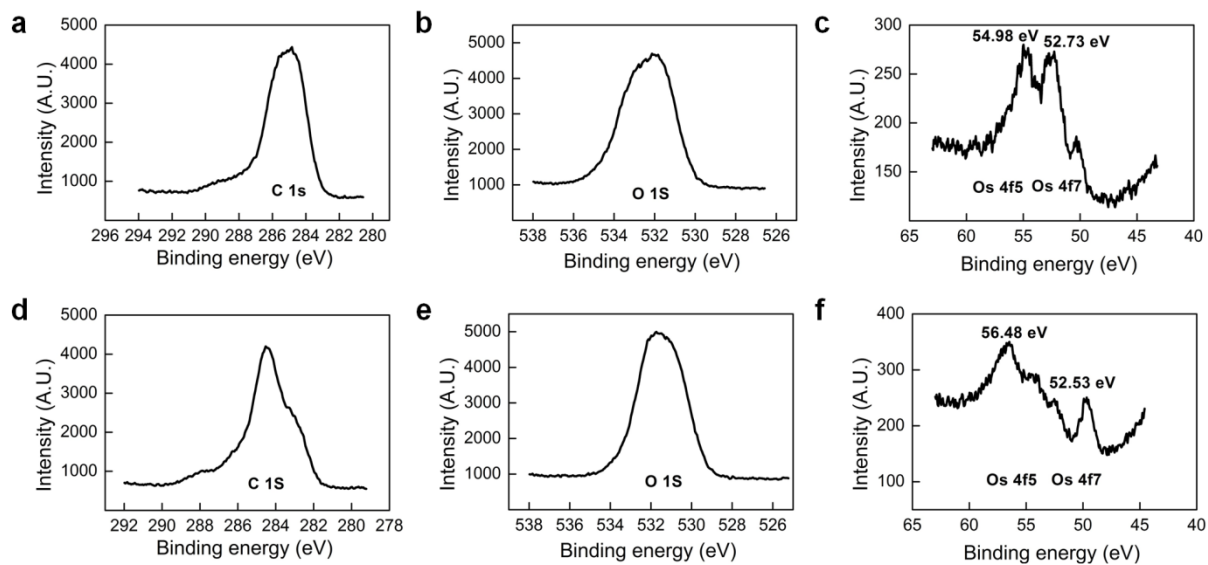
Supplementary Figure 1. SEM images showing (a) the surface of the PEDOT/MWCNT sheet used as yarn host and (b) the surface of the PEDOT/MWCNT sheet on which enzyme, redox mediator, and PEGDGE has been deposited as guests for making the biscrolled yarn cathode. The scale bars were 200 nm.



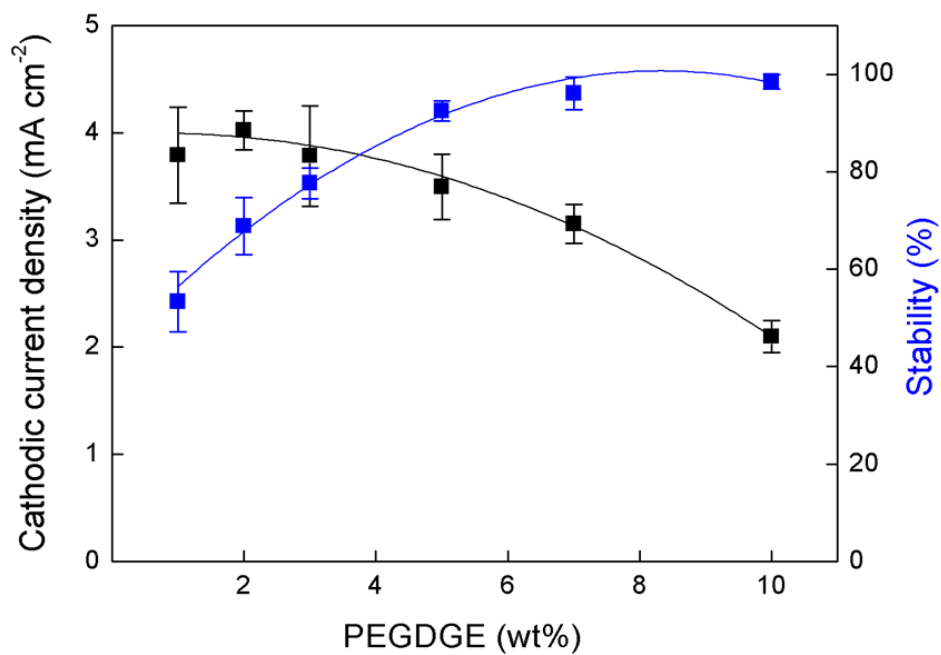
Supplementary Figure 2. Engineering tensile stress versus engineering tensile strain for non-modified guest-free PEDOT/MWCNT yarn (black line), the biscrolled anode yarn (blue line), and the biscrolled cathode yarn (red line) of the BFCs.



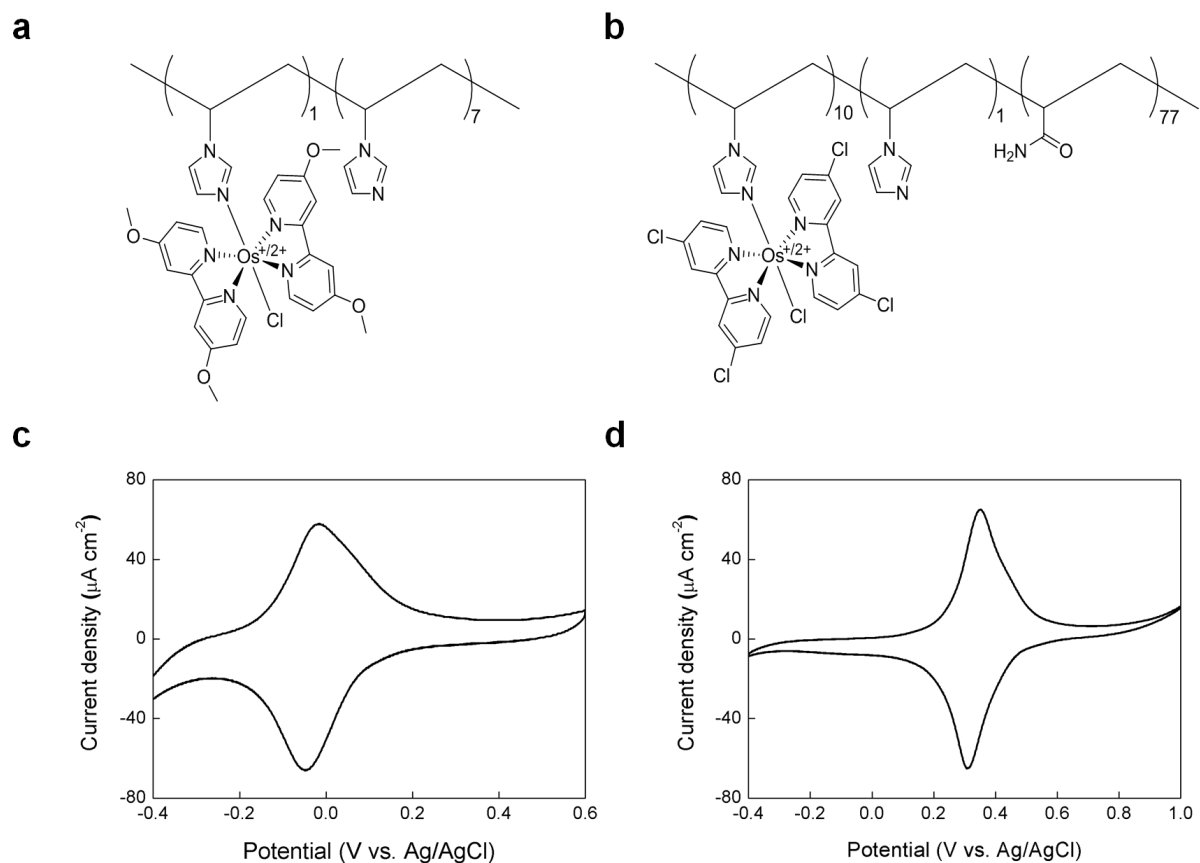
Supplementary Figure 3. Cyclic voltammetry for anode and cathode electrode in human serum condition at the scan rate of 5 mV s⁻¹.



Supplementary Figure 4. XPS spectra of anode electrode (**a-c**) and cathode electrode (**d-f**) to confirm the Os-containing guest materials in biscrolled-yarn electrode, composed of carbon (**a, d**), oxygen (**b, e**), and osmium (**c, f**).



Supplementary Figure 5. Optimal weight percentage of cross-linker (poly(ethylene glycol) diglycidyl ether, PEGDGE) depending on the maximum areal current density for a bisrolled cathode yarn. Data points represent average values of individual conditions and error bars indicate the s.d. from the mean value for three independent cases.



Supplementary Figure 6. Structures and redox peaks for anode and cathode redox mediators. **(a)** The structure of redox mediator I, which was used for the anode. **(b)** The structure of redox mediator II, which was used for the cathode. **(c)** Cyclic voltammety for the anode redox mediator (PVI-Os(dmo-bpy)₂Cl^{+/2+}) showing an average redox potential (E_0') at -38.5 mV vs. Ag/AgCl. **(d)** Cyclic voltammety for the cathode redox mediator (PAA-PVI-Os(dCl-bpy)₂Cl^{+/2+}) showing E_0' at 320.5 mV vs. Ag/AgCl. The scan rate was 100 mV s⁻¹.

Supplementary Note 1

Analysis of guests in BFC electrodes.

X-ray photoelectron spectroscopy (XPS, Thermo Fisher Scientific Co.) analysis was performed to confirm the electrode guest materials containing osmium in the modified biscrolled yarn electrode. As shown in **Supplementary Fig. 4**, the surface elemental composition on the surface of the electrode with respect to carbon 1s (284.53 eV) (**a,d**) and oxygen 1s (532.13 eV) (**b,e**) in the spectrum was determined. The XPS data show two peaks clearly for osmium 4f (anode 56.48 eV and 52.53eV; cathode 54.98 eV and 52.73eV) (**c,f**), which confirm the successful modification of osmium on the biscrolled yarn electrode. A single osmium compound with an electron binding energy of ca. 55 eV (4f electrons) dominates the spectrum, quite characteristic for hexavalent osmium¹. In anode and cathode electrodes, two compounds with similar binding energies were observed, possibly due to the formation of two different forms of osmium. In addition, the percentage of osmium in the biscrolled yarn BFC electrode was confirmed as a percentage of the total (C 1s, O 1s, and Os 4f: 72.66%, 27.32%, and 0.02% for anode, and 69.48%, 30.39%, and 0.13% for cathode, respectively).

Porosity and strength for the BFC electrodes.

Supplementary Fig. 1 shows the porous structures obtained for guest-free (**a**) and guest-coated (**b**) PEDOT/MWCNT sheet. **Supplementary Fig. 2** provides stress-strain curves for the anode and cathode yarns, which indicates the high mechanical strength needed for textile fabrication.

Supplementary Methods

Preparation of biscrolled yarn BFC electrodes.

Multiwalled carbon nanotube (MWCNT) sheets were drawn from ~400 μm high MWCNT forests that were grown by chemical vapor deposition (CVD) on iron-catalyst-coated silicon wafers using acetylene (C_2H_2) gas as the carbon precursor⁷. Iron(III) p-toluenesulfonate hexahydrate (Fe(III)PTS, M_w : 677.52), pyridine (anhydrous, 99.8%), 1-butanol ($\geq 99\%$), and 3,4 ethylenedioxythiophene (EDOT) monomer (97%) were purchased from Sigma-Aldrich (USA). For the polymerization of poly(3,4-ethylenedioxythiophene) (PEDOT), a 20 wt% solution of Fe(III)PTS in butanol (with 1.6 volume percent of subsequently added pyridine) was used. The Fe(III) acted as the oxidizing agent while PTS (or tosylate) acted as the anionic dopant. Fe(III)PTS/pyridine/butanol solutions containing 8 wt% of oxidant were made by diluting this stock solution with butanol. Eight wt% of oxidant solution was dropped over a two-layer carbon nanotube aerogel sheet stack, thereby delivering 70 μL of diluted solution for the 7 to 8 mm wide and 75 mm long carbon nanotube sheet stack. The carbon nanotube sheet stack was then dried at 60 $^\circ\text{C}$ for 20 minutes to evaporate the solvent. Densified Fe(III)PTS-containing sheets were thereby obtained.

Vapor phase polymerization (VPP) to coat the MWCNTs with PEDOT was accomplished by exposing these Fe(III)PTS-containing sheets to EDOT vapor in a VPP chamber for 1 hour at 60 $^\circ\text{C}$. After VPP, the PEDOT-containing MWCNT sheets were washed three times using distilled water. The average sheet thickness was ~102 nm for the host PEDOT/MWCNT sheet that was used as host for the biscrolled yarn. This two-layer PEDOT/MWCNT sheet contained 85 wt% PEDOT⁹.

Guest optimization.

The guests consist of mixtures of redox mediator, enzyme, and cross-linker. **Supplementary Fig. 6** shows the chemical structures (a, b) and redox peaks (c, d) for the anode and cathode redox mediators. The anodic redox mediator I was PVI-Os(dmo-bpy)₂Cl⁺²⁺, poly(N-vinylimidazole)-[Os(4,4'-dimethoxy-2,2'-bipyridine)₂Cl]⁺²⁺, and the cathodic redox mediator II was PAA-PVI-Os(dCl-bpy)₂Cl⁺²⁺, poly(acryl amide)-poly(N-vinylimidazole)-[Os(4,4'-dichloro-2,2'-bipyridine)₂]⁺²⁺. The maximum current density was measured as a function of enzyme concentration (**Fig. 2**) to obtain the selected enzyme concentration (41 wt%). The weight percentage of cross-linker (PEGDGE) was optimized by considering the maximum current density and stability, as shown in **Supplementary Fig. 5**.

Twist insertion process for guests coated PEDOT/MWCNT sheet.

A two-layer MWCNT sheet stack was supported by a glass slide during PEDOT coating by VPP and washing in distilled water using known process steps for PEDOT/MWCNT sheet fabrication⁹. After detaching the PEDOT/MWCNT sheet stack from the glass slide, the sheet stack was placed in an aqueous solution containing redox mediator, enzyme, and cross-linker, where it was held for 24 hours at 4 $^\circ\text{C}$. While still immersed in this solution and having one end fixed by tape, ~6,000 turns per meter of twist was then inserted into the sheet stack to make a biscrolled yarn. Symmetrical twist-insertion into the PEDOT/MWCNT sheet produced a dual-Archimedean yarn structure, as can be seen by the oppositely scrolled spinning cores (dark lines) on the two edges of the spinning wedge (**Supplementary Movie 1**).

Electrochemical measurements.

For the electrochemical measurements, the top end of a yarn electrode was cemented to a copper wire using commercial silver paste (ELCOAT P-100 from CANS, South Korea). The silver paste was allowed to dry and then insulated with an epoxy adhesive (5 Minute Epoxy S-208 from DEVCON, USA). Both the anodic yarn electrode (4.0-mm-long, 60- μ m-diameter) and the cathodic yarn electrode (4.8-mm-long, 50- μ m-diameter) had an active external surface area of 0.75 mm² for this part of the study.

A three-electrode electrochemical cell coupled to a CHI 600B potentiostat (USA) was used for cyclic voltammetry and chronoamperometry. A biscalloped yarn electrode was used as the working electrode, with Ag/AgCl reference electrode and a Pt mesh counter electrode. We used PBS buffer and human serum as electrolytes. PBS solution was freshly prepared before the experiments and human serum was purchased from Biological Specialty Corporation, USA. The measurements were conducted at 37 °C in an electrochemical cell containing 50 mL PBS solution (20 mmol L⁻¹ phosphate, 0.14 mol L⁻¹ NaCl) (pH: ~7.4), which was stirred at 500 rpm. Cyclic voltammetry for anode and cathode electrode in human serum was shown in **Supplementary Fig. 3**.

The anodic and cathodic current densities were determined by chronoamperometry at a constant voltage of 0.2 V for anode and 0.1 V for cathode, respectively, showing stable current density. The BFC power densities were determined by measuring current flowing through an external variable resistor (in the range of 10 Ω ~ 1 M Ω) to control cell potential.

Supplementary Reference

1. Herrmann W. A., *et al.* Polymer-bound osmium oxide catalysts. *J. Mol. Catal. A-Chem.* **120**, 197-205 (1997).