

Two-Ply Carbon Nanotube Fiber-Typed Enzymatic Biofuel Cell Implanted in Mice

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Abstract—Implantable devices have emerged as a promising industry. It is inevitable that these devices will require a power source to operate *in vivo*. Thus, to power implantable medical devices, biofuel cells (BFCs) that generate electricity using glucose without an external power supply have been considered. Although implantable BFCs have been developed for application *in vivo*, they are limited by their bulky electrodes and low power density. In the present study, we attempted to apply to living mice an implantable enzymatic BFC (EBFC) that was previously reported to be a high-power EBFC comprising carbon nanotube yarn electrodes. To improve their mechanical properties and for convenient implantation, the electrodes were coated with Nafion and twisted into a micro-sized, two-ply, one-body system. When the two-ply EBFC system was implanted in the abdominal cavity of mice, it provided a high-power density of 0.3 mW/cm². The two-ply EBFC system was injected through a needle using a syringe without surgery and the inflammatory response *in vivo* initially induced by the injection of the EBFC system was attenuated after 7 days, indicating the biocompatibility of the system *in vivo*.

Index Terms—Implantable enzymatic biofuel cell, multi-walled carbon nanotube (MWNT).

I. INTRODUCTION

IMPLANTABLE medical devices such as cardiac pacemakers, cochlear implants, neuro-stimulators, and insulin pumps have been developed for health problems that cannot be treated by surgery or drug therapy [1], [2]. a power source is necessary to operate these implantable medical devices. For this reason, commercial batteries are implanted with the devices. However, several critical problems are emerging in the use of commercial batteries as an energy source for implantable medical devices [2]–[4]. For instance, because of their limited life, batteries need to be replaced by surgery, which can be stressful to patients. Moreover, the volume and weight of a battery can be uncomfortable for patients in their daily living. Therefore, a power source optimized for conditions *in vivo* is an important challenge for further improvement of implantable medical devices. Numerous studies have been attempted to improve the power source for implantable devices, such as using energy harvesters [5], [6]. Mostly, these studies have used enzymatic biofuel cells (EBFCs), which can harvest electricity from biomolecular glucose supplied within the body. Thus, EBFCs have been investigated in various animals including insect, living snail and orange to generate a self-powered biomedical device system without an external energy supply [7]–[15]. However, bulk- [7], [16] or film- [17]–[19] shaped electrodes of implantable EBFCs thus far have a relatively large volume when implanted around tissues or organs, resulting in interrupted movement near the implanted site. Therefore, recent studies have focused on miniaturizing the volume of the EBFC while retaining efficient power [4], [11], [20], [21]. Here we report a miniaturized, one-body, two-ply, EBFC system and its successful implantation in the abdominal cavity of BALB/c mice. In the present study, we first made a high-powered and micro-sized cathode and anode yarn electrodes by twisting poly-(3,4-ethylenedioxythiophene) (PEDOT) coated multi-walled carbon nanotube (MWNT) sheets in an enzyme solution containing glucose oxide (GOx) for the anode and bilirubin oxide (BOD) for the cathode as reported previously with slight modification [18]. To implant the electrode *in vivo*, we further fabricated micro-sized cathode and anode yarn electrodes into a two-ply, one-body system. When our one-body EBFC was administered to the abdominal cavity of

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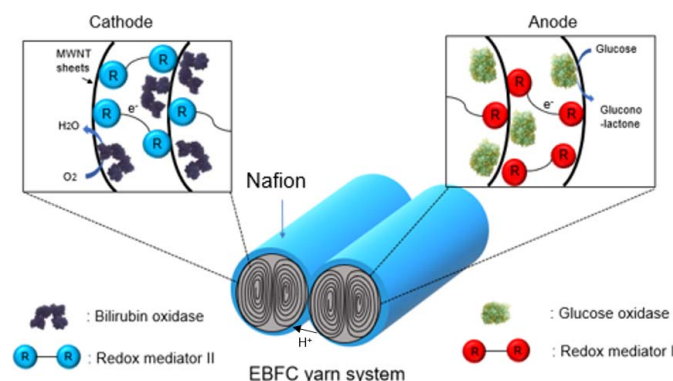


Fig. 1. Schematic illustration of EBFC yarn system. The EBFC yarn system formed bistructure that immobilized the enzymes and the redox mediators in the MWNT electrodes. glucose oxidase oxidizes glucose and the reaction generate the electron and the gluconolactone. The electron arrives in the MWNT electrodes through redox mediator I. In cathode, the electron transfer from the MWNT electrodes to bilirubin oxidase through redox mediator II and then, is reduced with oxygen and H^+ .

mice, it provided an areal power density of 0.3 mW/cm^2 and open circuit voltage (OCV) of 0.34 V . The implanted two-ply EBFC system evoked only a small and transient inflammatory reaction, suggesting that it is biocompatible.

II. EXPERIMENTAL

A. Materials

The host material of EBFC electrode is MWNT and guest materials of EBFC electrode were enzyme, redox mediator and cross-linker. MWNT forests consisting of $\sim 12 \text{ nm}$ diameter nanotubes were purchased from Lintec of America Inc (U53-HANYANG-SH215-09). 1-butanol ($\sim 99\%$ purity), pyridine (anhydrous, 99.8% purity), iron (III) p-toluenesulfonate hexahydrate (Fe(III)PTS) and 3,4-ethylenedioxythiophene (EDOT, 97% purity) were purchased from Sigma Aldrich. $\text{PVI-Os(dmo-bpy)}_2\text{Cl}^{+/2+}$, poly(N-vinylimidazo-le) $[\text{Os(4,4'-dimethoxy-2,2'-bipyridine)}_2\text{Cl}]^{+/2+}$ was used as anodic redox mediator. $\text{PAA-PVI-Os(dcl-bpy)}_2\text{Cl}^{+/2+}$, poly(acryl amide)-poly(N-vinylimid-azole)- $[\text{Os(4,4'-dichloro-2,2'-bipyridine)}_2]^{+/2+}$ was used as cathodic redox mediator [23]–[25]. In addition, glucose oxidase (GOx) from *Aspergillus niger* (180 U/mg) was used as anodic enzyme, which was purchased from Amano Enzyme Inc. (Nagoya, Japan). Meanwhile, the bilirubin oxidase (BOD) from *Myrothecium verrucaria* (1.2 U/mg) was used as cathodic enzyme, which also was purchased from Amano Enzyme Inc. (Nagoya, Japan). Poly (ethylene glycol) diglycidyl ether (PEGDGE) used to cross-link enzymes and redox mediators was purchased from Polysciences, Inc. Nafion ($5 \text{ wt\%}$) used to coat EBFC was purchased from Sigma Aldrich Corporation.

B. Fabrication of Two-Ply EBFC System

MWNT sheets drawn by 7-cm-long and 0.6-cm-wide stack of three layers were coated with poly (3,4-ethylene dioxothiophene) (PEDOT) using vapor phase polymerization as previously reported [26]. The EBFC yarn was twisted

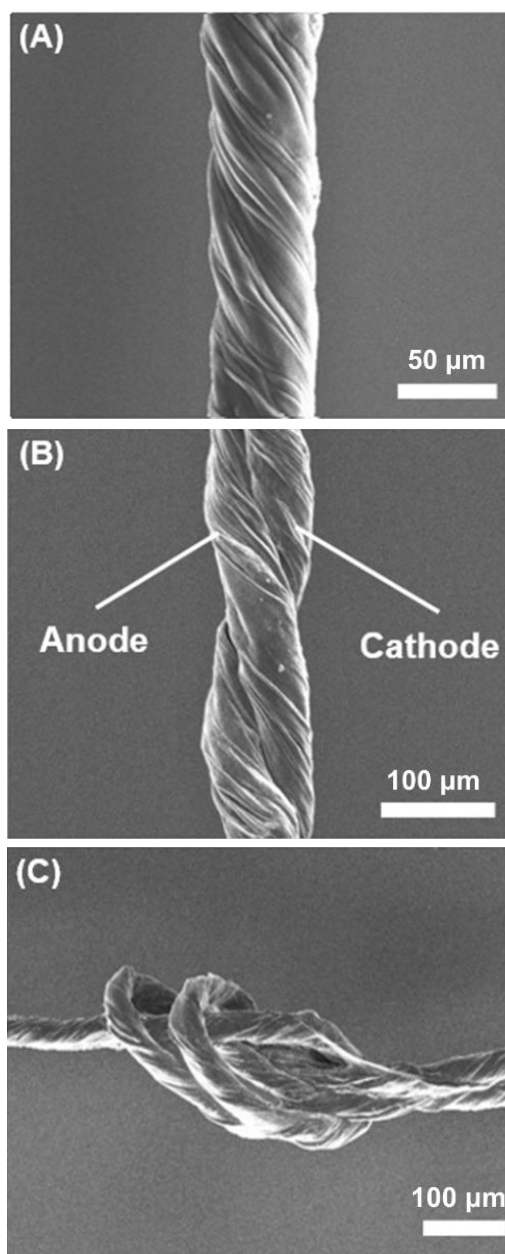


Fig. 2. Morphology of the implantable two-ply EBFC yarn system. (A) Scanning electron microscope (SEM) image of anode yarn coated with Nafion. (B) SEM image of the two-ply EBFC yarn system. (C) SEM image of a knotted two-ply EBFC yarn system.

in bath solution containing enzyme, redox mediator and crosslinker using the motor with ~ 2500 turns per meter of inserted twist per yarn length. The enzyme solution in anodic electrode consists of glucose oxidase ($41 \text{ wt\%}$), redox mediator I ($52 \text{ wt\%}$), and PEGDGE ($7 \text{ wt\%}$). The enzyme solution in cathodic electrode consists of bilirubin oxidase ($41 \text{ wt\%}$), redox mediator II ($54 \text{ wt\%}$), and PEGDGE ($5 \text{ wt\%}$). The average redox potential for the anode and cathode redox mediator is -38.5 mV and 320.5 mV (vs. Ag/AgCl), respectively [22]. After twist insertion, bistructured yarns were coated with Nafion ($5 \text{ wt\%}$) to prevent shortness of *in vivo* EBFC. Subsequently, anode and cathode EBFC yarns were twisted to two-ply. Normalization of the areal was calculated from the

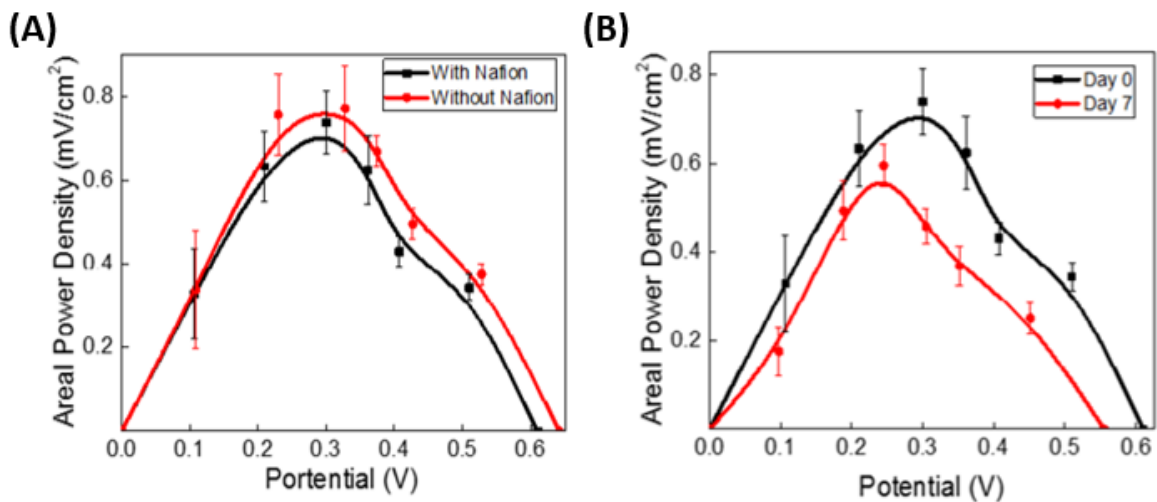


Fig. 3. The electrochemical performance of EBFC with Nafion in PBS buffer with 7mM glucose solution. (A) Comparison of the areal power density with operating Nafion-coated EBFC and operating uncoated EBFC. (B) Comparison of the areal power density for operating Nafion-coated EBFC with Day 0 and Day 7. The areal power density of EBFC were measured after 3 minutes of constant load. The error bars indicate standard deviation of five independent experiments.

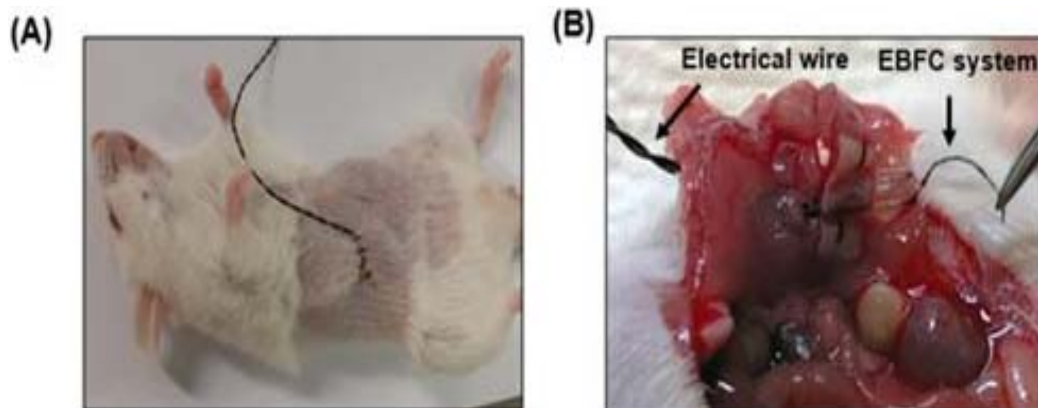


Fig. 4. (A) Mouse implanted with the two-ply EBFC system; the output wires are fixed at the inner surface of the mouse's abdomen. (B) Implanted two-ply EBFC system.

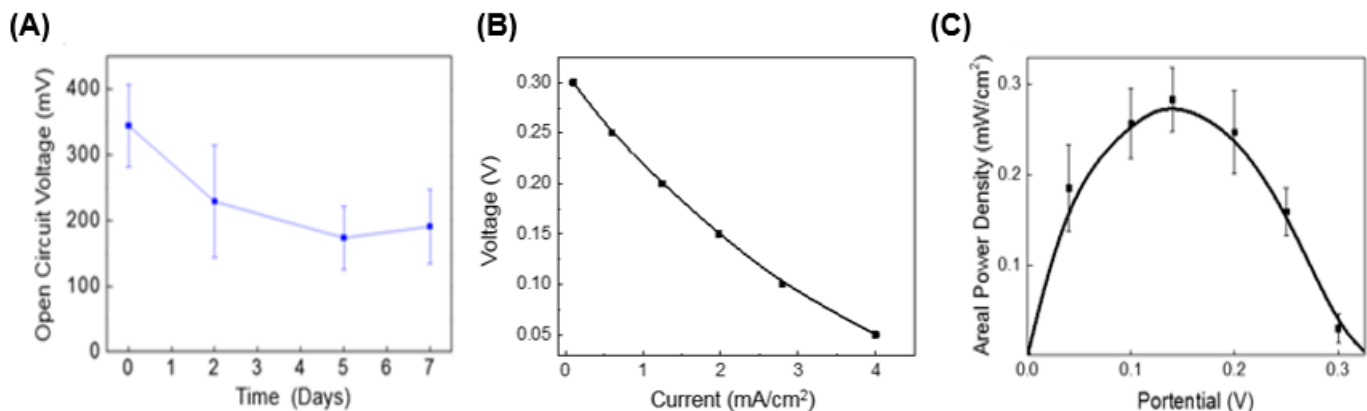


Fig. 5. Electrochemical performance of implanted the two-ply EBFC system. (A) Evolution of average OCV over time for 5 implanted EBFC system. (B) The polarization curve (V vs. I) of the two-ply EBFC system *in vivo*. (C) Areal power density as a function of potential for implanted EBFC system.

side surface of the cylinder without taking into account the roughness factor. Active external surface area of the EBFC system is 1.57 mm² in the study.

C. Characterizations and Electrochemical Measurements

Morphologies of EBFC yarns were characterized using scanning electron microscopy (Hitachi S4700).

All electrochemical performances of EBFC yarn system were determined by current flowing according to an external variable resistance. For *in vivo* analysis, the wired EBFC system was implanted through surgery.

D. Implantation Information

Animal care and experimental procedures were approved by the Institutional Animal Care and Use Community at

Samsung Medical Center (Permission number: 20151210001). The experiments were performed in accordance to their guiding principles (European Communities Council Directive L358-86/609/EEC). All protocols involving living animals were performed under license from the French Ministry of Agriculture (License number 38018 and 381141). The Committee on the Ethics of Animal Experiments of the Grenoble University.

E. Histological Analysis

Abdominal tissues were sampled at a location directly in contact with the EBFC surface. The tissues in direct contact with the EBFC surface were fixed with 4 % paraformaldehyde for 24 hours. The tissues were washed with PBS, and then embedded in paraffin for H&E staining. The tissue sections (4 μm) were stained with hematoxylin (BBC Biochemical, Mount Vernon, WA) for 5 minutes and eosin (BBC Biochemical, Mount Vernon, WA) for 1 minute. Stained tissue sections were examined under a light microscope [27].

III. RESULTS AND DISCUSSION

The EBFC yarn was fabricated by twisting PEDOT-coated MWNT sheets to immobilize enzymes and mediators on MWNT layers (Fig. 1). The diameter of EBFC yarn can be controlled by the amount of MWNT sheets and loading weight of guest materials. Because the EBFC system is easily exposed to mechanical stress due to the movement of the mice, structural stability is needed. To improve its mechanical properties for stable implantation, we used more CNT sheets for the implantable EBFC yarn than that used under in vitro conditions [22]. In addition, the biocompatible Nafion was coated on EBFC by 950 ng/cm to the electrical shorting of the anode and cathode electrodes (Fig. 2A) [28], [29]. The anode and cathode yarn electrodes were twisted into one-body system by two-ply to miniaturize its diameter to $\sim 70 \mu\text{m}$ (Fig. 2B). This one-body system not only reduces electro-chemical impedance, but also improves mechanical properties by reducing the distance between the anode and cathode [30]. Indeed, this EBFC yarn system exhibited sufficient flexibility and mechanical properties to make a knitted structure (Fig. 2C). Electrochemical performance of the Nafion-coated EBFC yarn system decreased about 9% for an operating EBFC yarn compared with that of uncoated EBFC yarn in PBS buffer with 7mM glucose solution (Fig. 3A). After 7 days, the areal power density for Nafion-coated EBFC system was 0.59 mW/cm^2 when operated on a fixed load, which indicates the reduction of 19% when compared to 0 day (Fig. 3B). To analyze the electrochemical performance in vivo of our two-ply EBFC system, it was surgically implanted into the abdominal cavity of mice that can fully contain the EBFC system (Fig. 4A). An electrical wire that was connected with the EBFC system was cosutured with the skin to prevent the movement of the EBFC yarn system in the abdominal cavity (Fig. 4B). After implantation, the mice were clothed with a commercial finger protection band to prevent damage to the wire from scratching and movement by the mice. Consequently, electrochemical performance was determined from five different EBFC yarn

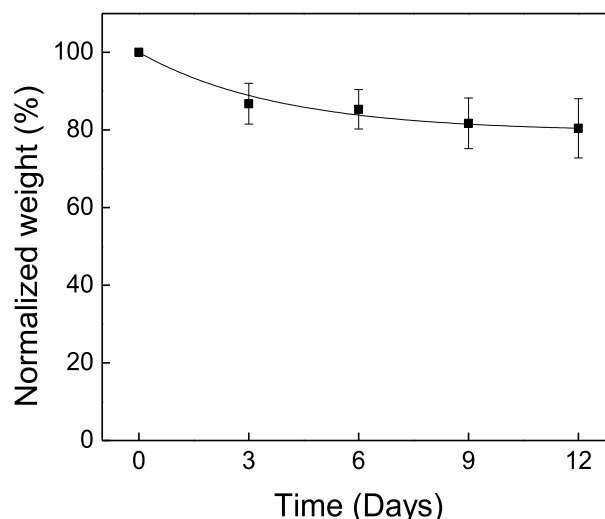


Fig. 6. The weight change graph of the enough dried EBFC yarn systems that was taken out after putting it in diluted water every 3 days for 12 days.

systems implanted in mice. We measured the OCV values of implanted EBFC yarn systems for 7 days (Fig. 5A). On the day of implantation, maximum OCV was measured in the range of 290–400 mV. The voltage decayed gradually and plateaued at 55% of the initial level from 5 days later. Indeed, The polarization curve (V vs. I) of the two-ply EBFC system implanted in mouse showed the open circuit voltage, 0.3 V and short circuit current, 4 mA/cm^2 (Fig. 5B). Fig. 5C indicates the variation of the areal power density as a function of the two-ply EBFC system voltage. When the applied voltage was between 0 and 0.34 V, the maximum power density shown was $298 \mu\text{W/cm}^2$ at 0.14 V. The areal power density of the EBFC yarn system was normalized to the surface of the two electrodes. Generally, fuels such as glucose and oxygen molecules should be easily transported into the BFC system for stable performance. By contrast, catalytic molecules such as enzymes and mediators should not leak from the BFC system. Given this point, our EBFC yarn system effectively prevented leakage of catalytic molecules, maintaining up to 80 % of its initial weight. This stability could be attributed to the layer-by-layer structure of MWNT that immobilizes catalytic molecules (Fig. 6). This result shows that our two-ply EBFC system exerts a higher areal power density than previous BFCs implanted in mammals (Table I). Because most implantable EBFCs thus far still have a bulk volume, surgery is needed to implant them in vivo. This surgical process mostly leads to severe inflammation around the implanted device. Because our EBFC has the advantage of its micro size, we attempted to inject it directly through a needle using a syringe and then investigated the inflammatory effect of the two-ply EBFC in the implanted mice. As shown in Fig. 7A, the two-ply EBFC system was administered to the abdominal cavity of BALB/c mice through a 26 G needle. Similar to conditions in vivo, we injected the EBFC system with 7 mM glucose-containing solution in the abdominal cavity of mice. We found the two-ply EBFC system was well implanted around tissue 7 days after administration as shown in Fig. 7B. To assess the inflamma-

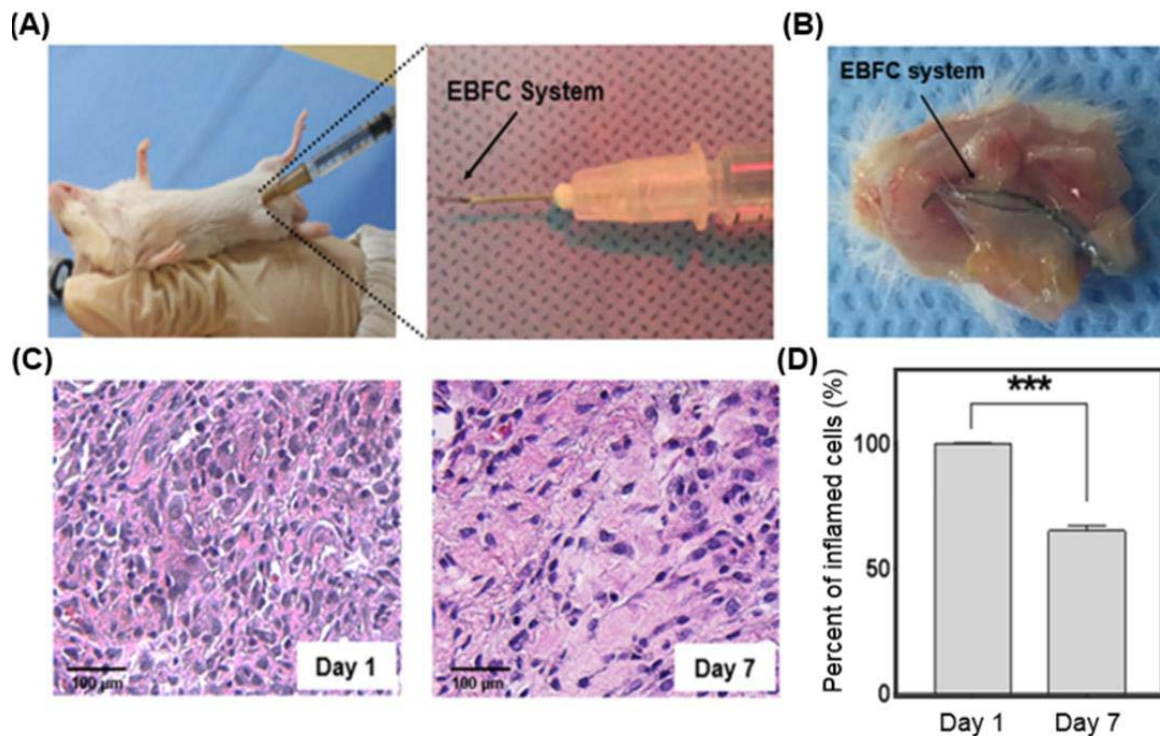


Fig. 7. (A) Optical image of the injection process of two-ply EBFC system into mouse fold without surgical operation via syringe (left). Optical image of two-ply EBFC system in the 26 G syringe needle (right). (B) optical images of the tissues surrounding the two-ply EBFC system after 7 days. (C) Images of tissue section around the BFC stained with H&E after 1 day (left) and 7 day (right). Purple dots represent Inflammatory cell. (D) Inflammation rate versus time in random three nearby tissues. The results are shown as mean $\pm$ SEM and student's t-test was used to decide statistical differences between mean of result about after 1 day and mean of result about after 7 days. Statistical significance was accepted for p-values < 0.001(***).

TABLE I
COMPARISON OF THE BFCs IMPLANTED IN MAMMALS

REF	Implantation site	Electrode	OCV (V)	Power ($\mu\text{W} / \text{cm}^2$)
4	Rabbit's ear vein	Carbon film with catheter	0.81	131
7	Rat's cremaster	MWCNT film	0.14	0.175
9	Rat's retroperitoneum	Graphite disk	0.275	24.4*
11	Rat's retroperitoneum	MWCNT pellet	0.57	193.5
16	Rat's brain cortex	AuNP wire	0.54	2
17	Rat's jugular vein	Carbon fiber	0.125	95
21	Rabbit's abdomen	MWCNT mass	0.42	55
OUR WORK	Mouse's abdominal cavity	MWCNT yarn	0.34	296

*: $\mu\text{W} / \text{mL}$

tory reaction of the tissues nearby the implant, we compared histology between 1 day (Fig. 7C. Left) and at 7 days (Fig. 7C. Right) after implantation. As shown in Fig. 7C, we found an increased initial inflammatory response due to the two-ply EBFC system around the implant 1 day after injection. This reaction is generally an acute immune response to foreign

substances in the body. Thus, the initial inflammation seen in the present study seems to be caused by a normal immune response to foreign material. The inflammatory response was attenuated after 7 days, indicating biocompatibility in vivo. Indeed, the number of inflammatory cells in the tissues implanted with the two-ply EBFC system for 7 days was

significantly attenuated to 66% of those observed 1 day after the injection (100%) (Fig. 7D, Student t test, *** $P < 0.001$). This finding suggests that our implanted EBFC system did not produce a harmful effect in nearby tissues.

IV. CONCLUSION

In conclusion, we successfully implanted our two-ply EBFC system into the abdominal cavity of mice without surgery. To our knowledge, this is the first time that such a system has been implanted into an animal smaller than a rat. The two-ply EBFC system provided electrical energy of $298 \mu\text{W}/\text{cm}^2$ *in vivo*. Our micro-sized EBFC system has the potential to be implanted by injection through a needle without the need for additional surgery. We suggest that a micro-sized, one-body system of yarn-type EBFCs could be applied to a variety of biomedical electrical devices, in a minimally invasive way without a membrane or packaging. Moreover, our EBFC system could be implanted as an energy source for powering microrobots to perform various functions such as drug delivery and treatments *in vivo*. However, further studies are warranted to improve the power and the stability *in vivo* for practical application.

AUTHOR CONTRIBUTIONS

D.Y.L., Y.B. P., and S.J.K. conceived the idea and designed the experiments; D.Y.L., Y.B. P., J.S.H. performed the experiments and analyzed data; J.-H. Y. contributed the animal experiment and histological analysis; D.Y.L., S.J.K., C.H.K., and Y.-W.J., wrote the manuscript; Y.B. P., J.-H. Y., J.S.H., S.M.K., R.H.B., and T.M.K. assisted with the writing of the manuscript; R. O. synthesized the CNT forests; Y.-B.C. and, H.-H. K synthesized mediators; All authors reviewed the manuscript.

ADDITIONAL INFORMATION

Competing Interests: The authors declare that they have no competing interests

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