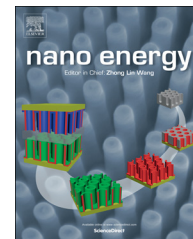




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RAPID COMMUNICATION

Lightweight, conductive hollow fibers from nature as sustainable electrode materials for microbial energy harvesting



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Abstract

Electrode materials with high specific surface area (area per volume or weight) and high conductivity are crucial for high power generation in microbial fuel cells (MFCs). In this paper, a novel hollow natural fiber template (kapok) is introduced to serve as the MFC anode. Advanced microscopy shows that the unique hollow structure doubles the anode active bacterial colonization surface area and for the first time provides double surfaces for microbial colonization through both internal and external surfaces. The high conductivity of hollow kapok fiber was directly achieved via carbonization. The power density of the anode comprised of carbonized hollow fibers (104.1 mW g^{-1}) is orders of magnitude higher than traditional solid fiber electrode (5.5 mW g^{-1}) on weight basis. These results demonstrated that with the same amount of material, generally measured in weights in engineering practice, this sustainable natural kapok fiber provides great advantages in improving MFC system performance and reducing cost.

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Introduction

As environmental problems, such as pollution, ozone depletion, fossil fuel shortage, and global warming, continue to intensify, a global emphasis on protecting the environment

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becomes increasingly urgent. Wood is an abundant and carbon neutral renewable resource. Using renewable biomaterials is an efficient way to maintain a clean and sustainable world. The kapok [*Ceibapentandra* (L.) Gaertn] trees supply an abundant quantity of round micro-tubular fibers with a thin cell wall thickness of 0.8–1.0 μm , a fiber diameter of 10–20 μm , and a length of several centimeters [1]. These natural organic tubes are widely available, low-cost, environmentally friendly, light-weight, and sustainable. Various tubes with a high length to diameter ratio have been successfully fabricated with kapok fibers as the template [2,3].

A microbial fuel cell (MFC) converts organic matter to electricity using electrochemically active bacteria (EAB) as the biocatalyst. A prospective application for MFCs is towards wastewater treatment, where the bacteria directly breaks down organic matter in wastewater and simultaneously generates clean energy. The extracellular electron transfer between EAB and anode surface occurs through direct contacts via pili and cytochromes or indirect electron transfer via mediators [4–9]. An ideal anode electrode should have a high surface area, high conductivity, biocompatibility, chemical stability, and three-dimensional (3D) macroporous structure to allow microbes to colonize or access for electron transfer [10–17]. Various conductive porous and three-dimensional materials have been investigated as MFC anodes, including carbon cloth [18], conductive textiles [10], carbon nanotube (CNT)/graphene coated sponges [12,19], layered corrugated carbon [20], electrospun and solution blown carbon fibers [21], and porous ceramic anode [22]. However, all the reported fibers are made of materials from graphite mines and have a solid structure with limited surface area that are difficult to be implemented in engineering systems due to the high weight, high cost, and unsustainable nature. Only a few current studies have developed sustainable and low cost anodes from natural materials for MFCs. Open structured macroporous bioanode was fabricated using a natural loofah sponge as the precursor material. Carbonization and polyaniline modification of the loofah sponge greatly increased its bacterial loading capacity and electron transfer, which favored much higher power production compared to traditional three-dimensional anodes [23]. Biochar manufactured with one-step pyrolysis process of lodgepole pine sawdust pellets or lodgepole pine woodchips was tested as a new anode material in two-chamber MFCs. The results showed satisfactory power outputs and it was 90% cheaper than granular activated carbon and graphite granules [24], but biochar anodes are still formed solid architecture which limits their specific surface area. Another study prepared an ordered 3D macroporous carbon anode for MFCs from the stem of kenaf (*Hibiscus cannabinus*), a natural plant; however, many channels in the structure are blocked by valves with only the open channels available for biofilm propagation [25].

The hollow fibers as electrode materials are distinctive from other porous and three-dimensional electrodes because all other electrodes are made of solid fibers that electrons can only travel on the outside surface, but the hollow structure fibers can provide both outside and inside surfaces for electron transfer. Furthermore, these hollow fibers are isolated from the natural plant kapok which makes the

hollow fiber material biocompatible, sustainable and low cost. In this study, we carbonized a low-cost and naturally forming hollow kapok fiber and used it as anode for the MFC, which provides a promising alternative to solid macroporous electrodes. The hollow structure of this conductive natural fiber doubles the active bacterial loading area with no increase in volume and weight, which offers larger surface areas for EAB to colonize and efficiently deliver electrons through both external and internal walls. The power generation with conductive kapok fiber anode from artificial wastewater was compared with traditional carbon cloth anode in the single-chamber air-cathode MFCs. The introduction of this high-performance anode from natural, low-cost, and sustainable electrodes for simultaneous waste management and renewable energy production provides a clean, cost-effective, and natural approach for sustainable community development.

Experiment

Preparation of conductive kapok anode

Natural kapok fibers were purchased from Bamboo Fiber (Bamboo Fiber Corp., US). They were washed thoroughly with ethanol and distilled water, then a free standing and porous kapok paper was prepared by vacuum filtration. The kapok paper was carbonized in a mixed gas atmosphere with 95% Ar and 5% H₂ in a tube furnace (Applied Test Systems, Inc.) from room temperature to 400 °C with a ramp rate of 30 °C h⁻¹, and further increased the temperature to 1100 °C with a ramp rate of 200 °C h⁻¹, then held the samples at 1100 °C for 2 h. After carbonization, the fiber was dispersed in distilled water and re-filtered to form a circular film with 40 mm diameter. The control sample without carbonization was fabricated directly from natural kapok fiber via vacuum filtration. The thickness of the kapok sample is $\sim 100 \mu\text{m}$.

MFC construction and operation

Single chamber cubic-shaped MFCs were constructed as previously described [26]. The empty volume of each MFC was 28 mL. Air cathodes (projected area of 4.5 cm²) made by applying one carbon base layer, four PTFE diffusion layers and one Pt/C (0.5 mg cm⁻²) catalyst layer on the 30% wet-proofed carbon cloth (Fuel Cell Earth LLC, MA, USA) were used in all the experiments [27]. The carbonized kapok anodes (kapok_c) and non-carbonized kapok anodes (kapok_nc) were prepared as described above. Plain carbon cloth (CC, Fuel Cell Earth LLC, MA, USA), commonly used as anode material, was used as a control sample for kapok anodes.

MFCs were inoculated using anaerobic sludge obtained from Littleton/Englewood Wastewater Treatment Plant (Englewood, CO). Medium solution was prepared containing 1.24 g L⁻¹ CH₃COONa, 0.31 g L⁻¹ NH₄Cl, 0.13 g L⁻¹ KCl, 2.452 g L⁻¹ NaH₂PO₄ · H₂O, 4.576 g L⁻¹ Na₂HPO₄, 12.5 mL L⁻¹ mineral solution, and 5 mL L⁻¹ vitamin solution [28]. All MFCs were operated in fed-batch mode at room temperature. Fresh medium solution was refilled when the voltage dropped below 10 mV with an external resistor of 1000 Ω .

Analyses

The MFC voltages and electrode potentials were recorded over 10 min intervals using a data acquisition system (Keithley Instrument, OH, US). The anode potentials and cathode potentials were measured against a Ag/AgCl reference electrode (RE-5B, Bioanalysis) at a distance of 1 cm to the anode in the reactor. Polarization curves were obtained by Linear Sweep Voltammetry (LSV) at a scan rate of 0.1 mV s^{-1} using a potentiostat (PC4/300, Gamry Instruments, NJ). Electrochemical Impedance Spectroscopy (EIS) was conducted using the same potentiostat to measure the internal resistance at the scan range from 10^5 Hz to 0.005 Hz with a small sinusoidal perturbation of $\pm 10 \text{ mV}$. In both LSV and EIS tests, the anode was used as the working electrode and the cathode was used as the counter electrode and reference electrode. Chemical oxygen demand (COD) was measured using a standard colorimetric method (Hach Company, CO).

COD removal efficiency is the ratio of total COD removed at the end of the batch to influent COD in percentage. The output power (P) of MFC was calculated by $P=UI$, where U is the voltage across the MFC anode and cathode and I is the MFC output current. Current density was normalized by the projected area of the anode (4.5 cm^2). Power density was normalized by the projected area, reactor volume (28 mL) and the mass of the anode.

Non-carbonized and carbonized kapok and carbon cloth samples were characterized with Hitachi SU-70 FESEM field effect scanning electron microscopy (SEM), performed using a Jeol JXA 840A system (Jeol Instruments, Tokyo, Japan) running at 5-10 keV. Samples were fixed overnight by Karnovsky's fixative (Electron Microscopy Sciences, CA, USA) at 4°C , washed three times in a pH 7.2 phosphate buffer, and then dehydrated stepwise in a series of ethanol/water solutions with increasing ethanol concentration of 50%, 70%, 80%, 90%, and 100%. Samples were then kept in a desiccator prior to carbon coating and SEM observation. The fiber surface area was measured with a Micromeritics TriStar II 3020 Porosimeter Test Station. The surface of kapok fiber after carbonization is $820 \text{ m}^2 \text{ g}^{-1}$.

Results and discussions

Kapok trees as shown in Figure 1(a) are widespread in tropical areas. They are 60-70 m tall and their trunk expands to 3 m in diameter with palm like leaves. Adult kapok trees produce several hundred seed pods with black seeds and fibers that are shades of yellow Figure 1(b). The fluffy light fibers extracted from the seed pods of the kapok trees are used in pillows, life preservers, sleeping bags, and insulation. These fibers are also recently investigated as an oil absorbent and sound absorber [29-32]. This study took hollow kapok fibers as an anode in the MFC. High surface area is critical to increase the microbial loading and improve the power density for large scale MFCs [33,34]. Due to the open 3D hollow structure, the microbes colonize both the inner and outer surfaces of the fiber wall, as illustrated in Figure 1(c). The biofilm mass loading is doubled within the same electrode volume. The generated electrons transport along both the internal and external surfaces of the fiber wall, thus the double surfaces for

microbial colonization dramatically increase the power density. Meanwhile, since the kapok fiber is produced in nature, the fiber has good biocompatibility, which benefits the contact between microbes and the anode to enhance the electron transfer efficiency. These natural, lightweight, and cost efficient hollow fibers are promising electrodes for MFCs.

Carbonization of biomass is a promising strategy for carbon materials from sustainable resources [35]. The kapok fiber is a natural biocomposite composed mainly of cellulose and lignin. Figure 2(a) and (b) show the fiber morphologies before carbonization. Figure 2(a) illustrates the kapok fiber has a round shape and smooth surface with a diameter of 10-20 μm . In Figure 2(b), the thin wall of kapok fibers is well defined. The fiber has a large empty lumen, which provides enough space to host the microbial growth and substrate reservation within the lumen. Since 77 v% of the fiber is hollow, the density of kapok fiber is as low as 0.384 g cm^{-3} [36,37], which is much lighter than the solid fiber; for example, the density of carbon cloth is 1.75 g cm^{-3} [38]. Weight is a huge concern when designing portable systems for truck delivery, because it can be very heavy when filled with water. The lightweight of conductive fibers in MFCs will reduce the cost in transportation and realize large-scale practical applications. The lightweight of kapok is therefore a great benefit for fuel cells of any size.

Kapok fibers function as a microbial growth scaffold and more importantly facilitate the electron transfer in the MFC. The conductivity of kapok fiber is thus important, but the pristine fibers are not conductive. The threat of an energy shortage continues to drive the pursuit of a facile, simple, and environmentally friendly method to prepare advanced functional materials. There are a lot of studies on making conductive carbonaceous materials from earth abundant biomass via carbonization [35,39]. We carbonized kapok fiber in a mixed gas, see Experiment section. The pyrolysis process of cellulose mainly includes dehydration and depolymerization. The reason for using a slow ramp rate before 400°C is to reduce the carbohydrate material burning loss thereby improving yield and properties of obtained carbon fiber [39]. Figure 2(c and d) shows the fiber morphology after carbonization. From the images we can see the hollow structure is well preserved after carbonization, and the fiber walls become thinner. More SEM images of kapok fiber after carbonization are presented in Supplemental material Figure S1. In order to measure the fiber conductivity, we fabricated two-probe devices with our hollow carbon fiber on glass substrates. Uniform fibers were fixed and contacted by silver paste (Figure 2(e)) and the electrical characterization was carried out in a vacuum using a shielded probe station. We observed a linear I - V curve with a source drain bias V_{ds} reaching $\sim 1 \text{ V}$ (Figure 2(f)). Such ohmic I_{ds} - V_{ds} behavior indicates that the Schottky barriers at the contacts can be excluded. The more fiber morphology and I - V curve are presented in Supplemental material (Figures S2 and S3). The volume resistivity of $\rho \sim 140 \pm 30 \Omega \mu\text{m}$ represents the upper limit due to the contribution from contact resistance, from which we can see the kapok fiber has a good conductivity after carbonization.

After acclimation, repeatable cycles of voltage generation were obtained from all the reactors as shown in Figure 3(a). The voltage generation over a $1 \text{ k}\Omega$ external resistor in the kapok_c reactor is $514.9 \pm 3.9 \text{ mV}$, which

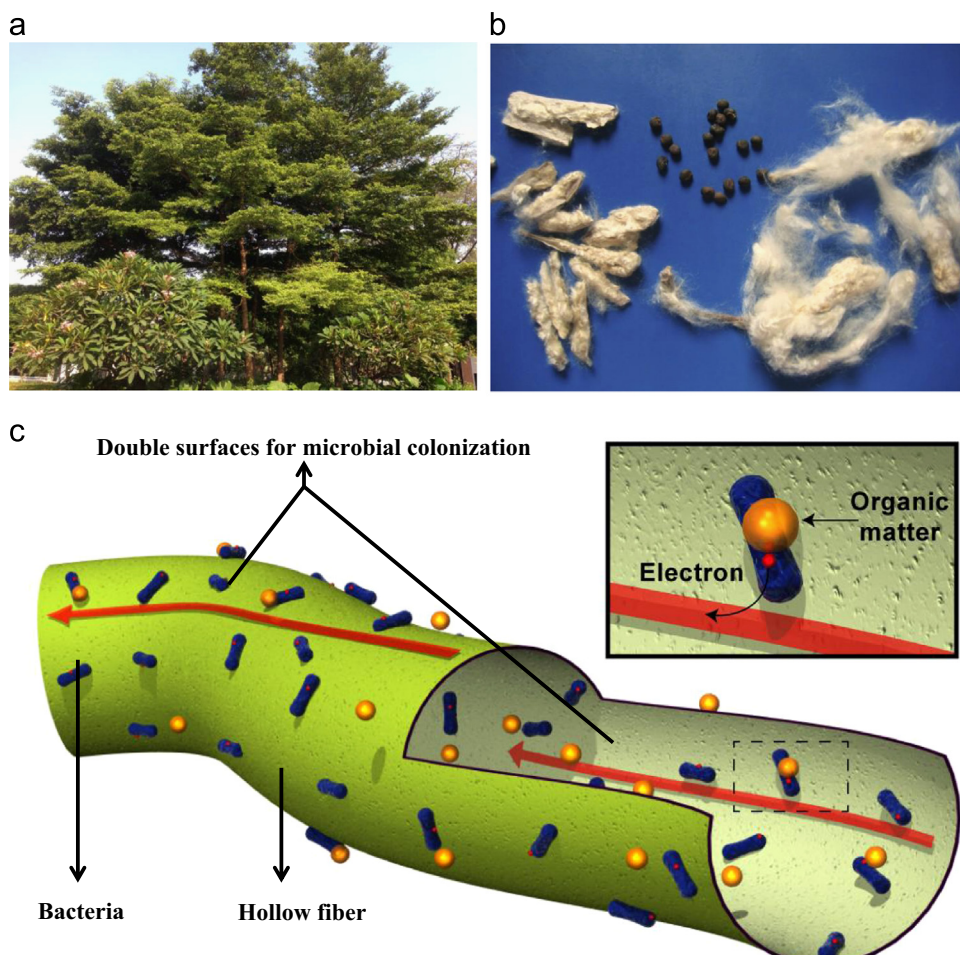


Figure 1 (a) A picture of the kapok tree. (b) An image of the kapok fibers and the seeds from the kapok seed pods. (c) The schematic shows the hollow kapok fiber as the electrode support for the microbial growth and provides double surfaces for microbial colonization through both internal and external surfaces. The kapok fiber applied in the MFC is post-carbonization with an excellent conductivity. Note that bacteria can grow both inside and outside the hollow fiber, maximizing the direct contact of bacteria with conductive fiber surface.

is 9.3% higher than that in the kapok_nc reactor (470.9 ± 4.8 mV) and comparable to that in the CC reactor (520.0 ± 2.6 mV). COD removal efficiencies were comparable in all the reactors, with $92.9 \pm 2.1\%$ for kapok_c, $92.5 \pm 1.0\%$ for kapok_nc, and $94.7 \pm 0.8\%$ for CC. Under stable performance, polarization curves were obtained by LSV and then power densities were calculated (Figure 3(b)). The maximum power density generated in the kapok_c reactor and normalized by the projected anode surface area is 1738.1 mW m^{-2} , which is 30.9% higher than that in the kapok_nc reactor (1328.1 mW m^{-2}) and comparable to that in the CC reactor (1689.8 mW m^{-2}). When normalized by reactor volume, the power densities of the kapok_c reactor (27.9 W m^{-3}) and the CC reactor (27.1 W m^{-3}) are comparable, which are 27.2% higher than that of the kapok_nc reactor (21.3 W m^{-3}). Due to the hollow structure of kapok fiber, the mass of the kapok anodes (kapok_c and kapok_nc) is only 7.5 mg, which is much lighter than the CC anode (139.1 mg). Normalizing the power densities by mass, kapok_c and kapok_nc generated powers of 104.1 mW g^{-1} and 79.7 mW g^{-1} , respectively, but CC only produced 5.5 mW g^{-1} (Figure 3(c)). It should be noted that the power

generation of kapok_nc MFC was due to the contribution of CNTs on the electrode. CNTs are mixed with the kapok fiber in order to improve the mechanical strength of the kapok film; otherwise it is hard to create a structure to serve as the electrode. Carbonization of non-conductive kapok fibers indeed improves the performance of the MFC, with power densities increase by 30.9%, 31.0%, and 30.6%, respectively, if normalized by electrode projected surface area, reactor volume and electrode mass. Figure 3(d) shows the anode potentials and cathode potentials measured during LSV tests. All the cathode potentials are apparently similar at each current density in the three reactors; the anode potentials are also similar in the kapok_c reactor and the CC reactor, but the anode potentials in the kapok_nc reactor are higher than that in the other two reactors. This result confirms that the differences of power generation in the three reactors are due to different anode performances but not cathode performances. Figure 3(e) shows the total resistance measured by an EIS test for MFCs with three different anode materials. The intersection of the curve with the x-axis indicates the ohmic resistance, and the diameter of the semicircle presents polarization resistance

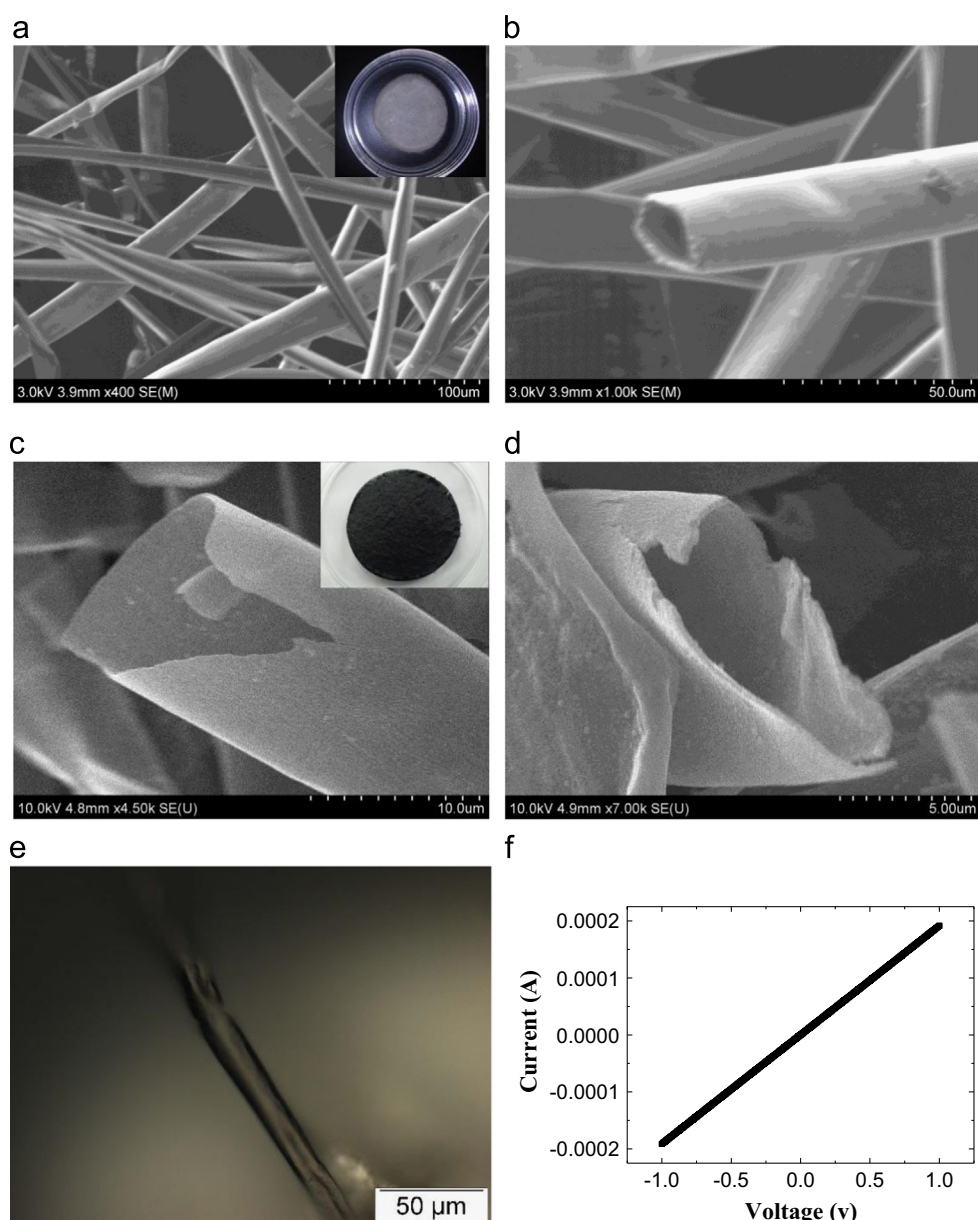


Figure 2 (a) An SEM image of the surface morphology of pristine kapok fibers. Inset image is a kapok paper fabricated through papermaking process. (b) An SEM image of the natural hollow tube. Both (a) and (b) are before carbonization. (c) and (d) are SEM images of the kapok fiber morphology after carbonization. Inset picture in (c) is an image of kapok paper after carbonization. (e) A microscope image of an uniform single fiber fixed and contacted by silver paste for the conductivity test. (f) The I - V curve of a single kapok fiber in (e).

or charge transfer resistance [40]. The ohmic resistances of the reactors with kapok_c or kapok_nc are about 72Ω and 68Ω , respectively, which are a little higher than that of CC (65Ω). Because of the same reactor configuration and electrolyte solutions, the ohmic resistances of all the reactors are generally comparable. The ohmic resistances of the reactor with kapok electrode are a little higher than the reactor with CC electrode which may be due to the relatively low conductivity of uncarbonized kapok materials and the loose connection between carbonized kapok electrode and the titanium wire which is used to link with the outside circuit. The polarization resistances of kapok_c and CC are both very small; while the polarization

resistance of kapok_nc was as high as 20Ω . Charge-transfer resistance is closely relevant to the properties of electrode surface which determines the kinetics of electron transfer. Both CC and kapok_c electrodes have small charge-transfer resistances which can well assist electron transfer, however, the high charge-transfer resistance of kapok_nc is detrimental to electron transfer due to the high resistance on the surface of kapok_nc electrode. The total resistance of the reactor includes both ohmic resistance and charge-transfer resistance, which is correlated to the performance of MFC reactors. Power productions of the reactors with CC or kapok_c are comparable since their resistances are also similar. The reactor with the kapok_nc

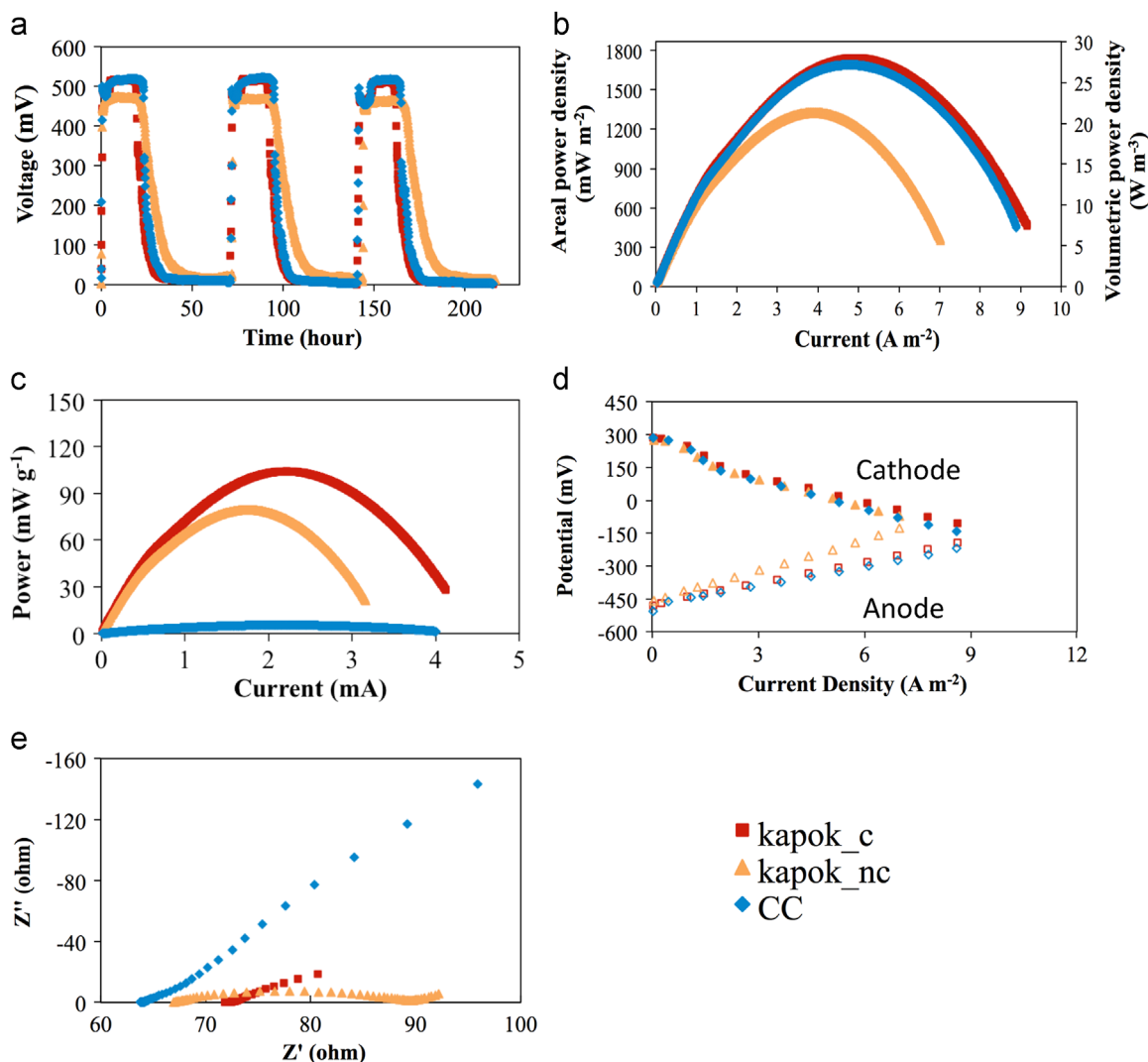


Figure 3 A demonstration of energy production in MFCs using carbonized kapok hollow fibers (kapok_c) as the anode with comparisons to uncarbonized kapok hollow fibers (kapok_nc) and traditional carbon cloth anodes (CC). (a) Repeating voltage generations under a 1 k Ω external resistor. (b) Power production curves obtained from linear sweep voltammetry (LSV) test clearly show the maximum power density at the peak. Power densities are normalized to the projected anode surface area and reactor volume. (c) Power densities are normalized to anode mass. (d) Anode and cathode potentials measured during LSV test. (e) Electrochemical impedance spectroscopy (EIS) shows the total resistance of MFC with three different anode materials.

anode generated the lowest power because the high resistance enhances the energy loss and decreases the power output. Since the surface conductivity is low because there is no carbonization treatment for kapok_nc, which is not favorable for electron transfer. Although the MFC with kapok_c anode has a higher total resistance than the MFC with CC anode, kapok_c can generate, at least, comparable power density as CC, which indicates that the hollow structure can utilize both the inside surface and outside surface to accelerate electron transfer, but the solid fiber anode, such as CC, can only transfer electrons through the outside surface. The kapok hollow fiber anodes in this study are the first try, and their manufacturing procedures have not been optimized yet. Therefore more efficient and conductive kapok hollow fibers can be manufactured, and the power production may be further enhanced following an optimized manufacturing process.

The hollow conductive fiber provides a dual microbial-anode interface. The microbial growth is evident on the internal and external wall of the fiber, which not only increases the surface area and biofilm loading, but more importantly provides double surfaces for microbial colonization on the anode. Solid surfaces of carbon cloth fibers are incapable of exhibiting this behavior (Figure 4(e) and (f)). The thickness of the biofilm depends on the oxygen and substrate concentration [41]. The anode chamber of the MFC reactors is in an anaerobic condition, which means the biofilm on the anode grows slowly in the limited oxygen environment. The COD of the substrate used in this study is about 1000 mg L⁻¹, which is double that of typical domestic wastewater with a COD of 500 mg L⁻¹ [42]. Biofilm cannot grow as fast as that in the high concentrated substrate. SEM images also clearly show that the diameter of the hollow fiber (μm) is much bigger than bacteria sizes (0.5-1 μm). In

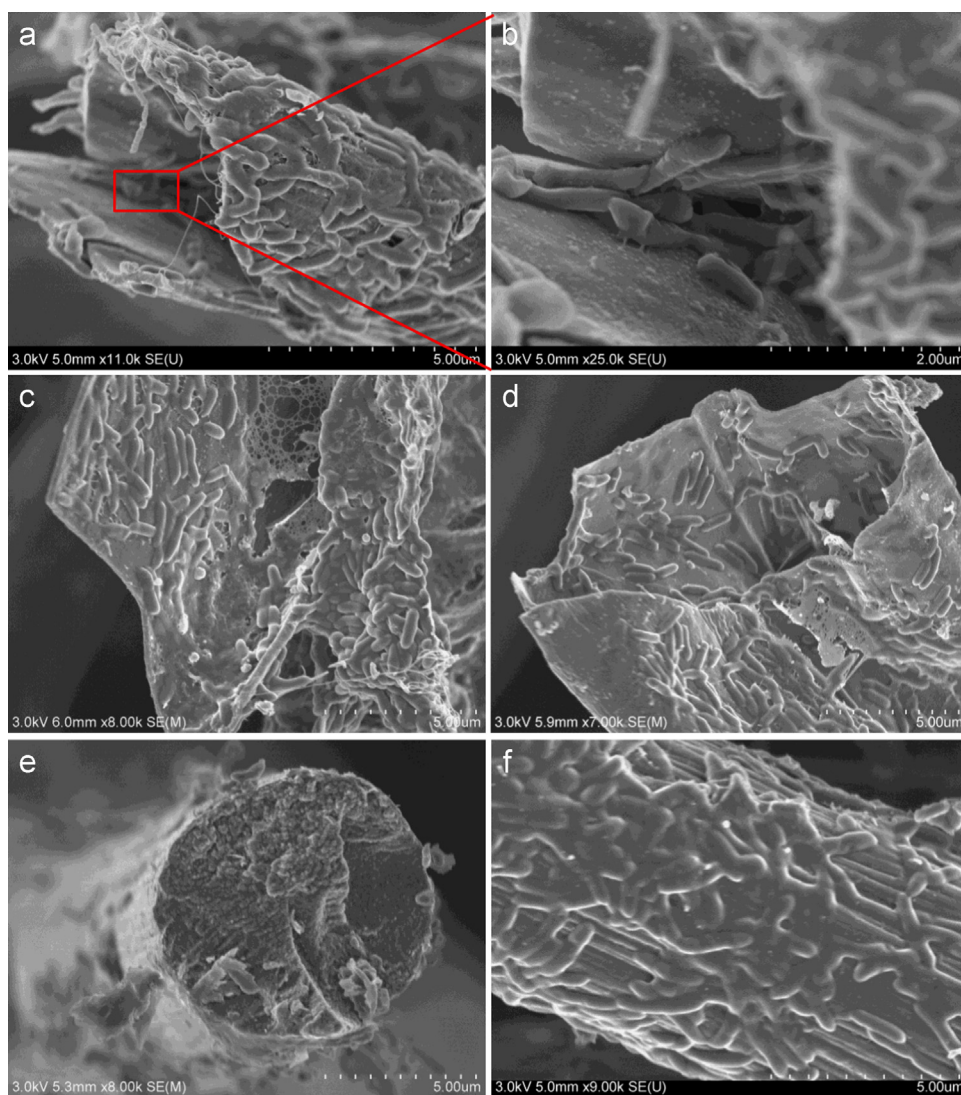


Figure 4 (a)-(d) SEM images of the microbial growth on zthe kapok fiber. (b) Magnified SEM image of red area in (a). These SEM images clearly confirm that bacteria grow on both sides of the conductive hollow fiber wall. (e) and (f) SEM images of the microbial growth on the carbon cloth fiber. More SEM images of bacterial colonization on the kapok fiber and carbon cloth are presented in Supplemental material [Figure S4](#).

the three-month long lab testing of the hollow fiber anodes in MFCs, no clogging was found and the performances of the reactors were stable. More studies are still needed to evaluate long-term performances of the hollow kapok fiber. For the solid carbon cloth fibers, the bacteria can only colonize on the outer surface of solid carbon cloth fibers ([Figure 4\(e\)](#) and ([f](#))), which limits the bacteria loading and has a much larger density than the hollow fiber. Note that the mass density of carbonized kapok fibers in this study is 59.7 mg cm^{-3} .

Conclusion

This study introduces an open-structured, hollow 3D macroporous anode formed from natural kapok fibers. The hollow structure doubles the microbe-anode interface while maintaining the same volume of solid fibers thus significantly increases the area for microbial acclimation and electron

transfer. The lightweight of such natural fiber provides a great advantage in large-scale engineering applications and modular system development, as surface/weight ratio is a major challenge for scale-up now. The direct carbonization of sustainable cellulosic fibers offers a facile and simple way to prepare conductive carbon fibers [43]. Normalizing with respect to the projected anode surface area and reactor volume, the power density production from the MFC using this carbonized low cost hollow-fiber structure anode (1738.1 mW m^{-2} or 27.9 W m^{-3}) is comparable to the expensive solid-fiber structured carbon cloth (1689.8 mW m^{-2} or 27.1 W m^{-3}). Normalized power density by mass, the power density production using the lightweight carbonized kapok anode is 104.1 mW g^{-1} , which is about 20 times as that produced by carbon cloth anodes (5.5 mW g^{-1}). The hollow natural fibers provide a new concept for designing high-power, lightweight, and sustainable MFC anodes with earth abundant and low cost materials that are greatly amenable to engineering applications.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.nanoen.2014.08.014>.

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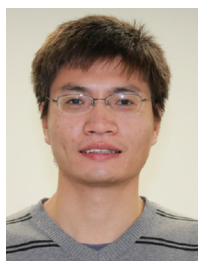
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