**Towards high power output of scaled-up benthic microbial fuel cells (BMFCs) using multiple
electron collectors**

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16 Abstract

17 This study aimed at achieving high power output of benthic microbial fuel cells (BMFCs) with novel
18 geometric anode setups (inverted tube granular activated charcoal (IT-GAC) and carbon cloth roll
19 (CCR)) and multiple anodes/electron collectors. The lab-scale tests showed the power density of IT-
20 GAC and CCR anodes achieved at 2.92 and 2.55 W m^{-2} , the highest value ever reported in BMFCs. The
21 power density of BMFCs substantially increased with electron collector number (titanium rods) in
22 anodes. The connection of multiple electron collectors with multiple cathodes had much higher total
23 voltage/current output than that with single cathode. The possibility of maintaining high power density
24 at scaled-up BMFCs was explored by arranging multiple anodes in sediment. The compact configuration
25 of multiple CCR anodes contacting each other did not deteriorate the performance of individual anodes,
26 showing the feasibility of maximizing anode numbers per sediment footprint and achieving high power
27 output. Multiple IT-GAC and CCR anodes with multiple collectors effectively utilized sediment at both
28 horizontal and vertical directions and enhanced electron collection efficiency. This study demonstrated
29 that bacterial adhesion and electron collection should be optimized on small anodes in order to maintain
30 high power density and achieve high power output in the scaled-up BMFCs.

31 **Keyword:** Benthic microbial fuel cell, power output, power density, electron collectors, granular
32 activated charcoal, carbon cloth rolls.

33

34 **Graphic Abstract:**

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36

37 1. Introduction

38 Rapid development of marine exploration (e.g. ecosystem observation, pollution detection,
39 disaster prevention) poses critical requirements of reliable and scalable power supply for subsea sensor
40 networks (Proakis et al., 2001; Akyildiz et al., 2005; Heidemann et al., 2006; Heidemann et al., 2012).
41 Lithium battery has been widely used as the subsea power source, but its deployment and replacement is
42 costly (Heidemann et al., 2006). In recent years, benthic microbial fuel cell (BMFC) has been explored
43 as an alternate power source for subsea sensors (Reimers et al., 2001, 2006). Compared with traditional
44 battery, BMFCs utilize the endless sources of organic substances and microorganisms in ocean sediment
45 as the anode and oxygen in ocean water as cathode. Such a system could provide a long term and stable
46 power supply for marine sensor systems and potentially reduce the costs for device deployment,
47 maintenance, and replacement.

48 However, one major problem of BMFCs is low power output that fails to meet the requirements
49 to run these marine systems (Tender et al., 2002; Nielsen et al., 2008; Tender et al., 2008; Chadwick et
50 al., 2010; Gong et al., 2011; Hsu et al., 2013). Until now, most BMFC systems only use single electrode
51 configurations (with one anode and/or cathode) (Reimers et al., 2001, 2006; Tender et al., 2002; He et
52 al., 2007; Nielsen et al., 2007; Donovan et al., 2008; Nielsen et al., 2008; Tender et al., 2008; Chadwick
53 et al., 2010; Gong et al., 2011; Hsu et al., 2013). A semi-chambered BMFC (footprint area: 0.25 m²,
54 foiled anode carbon brush length: 4 m) achieved an average power density of 44 mW.m⁻² based on
55 anode footprint area (Gong et al., 2011). A BMFC with flat carbon fabric anodes was scaled up from 25
56 cm² to 12 m², but the power density per anode footprint decreased from 23 mW.m⁻² to 2 mW.m⁻² (Hsu et
57 al., 2013). BMFCs with a single anode/cathode pair have three major problems. First, the inherent low
58 voltage difference (theoretical value: 1.2 V) of the anode and cathode in a MFC could not meet the
59 voltage/current/power requirements of most subsea sensors (e.g. 3.3~28 V and 49.5~560 mW) (Logan et

60 al., 2006; Nielsen et al., 2008). Second, single anode/cathode pair could easily malfunction under
61 bioturbation (e.g. oxygen leaking caused by invasion of aquatic life), not only oxygenating the anode but
62 possibly damaging the BMFC system, thus jeopardizing its ability to provide a stable long-term power
63 supply (Karra et al., 2013; Liu et al., 2015). Third, although sediment has a large amount of
64 microorganisms and nutrients that can produce electrons via MFCs, it is not a good electronic conductor,
65 meaning that anodes have to occupy the whole sediment footprint in order to obtain the electrons
66 generated by bacteria. But the potential difference between anode and cathode is fixed (~1.2V) for
67 BMFCs, leading to a tremendous drop of power density in scaled-up BMFCs. Unlike alkaline batteries
68 filled with high conductive metal powder capable of maintaining power density during scaling up, the
69 power density of the scaled-up MFCs filled with low conductive sediment dropped in magnitude orders
70 (Dewan et al., 2008, 2010; Hsu et al., 2013). Therefore, in order to mitigate the electron transfer
71 resistance and enhance power output at scale-up BMFCs, deploying multiple electrodes compactly with
72 small footprint in sediment appears to be the most feasible approach. A new configuration of multiple
73 anodes/cathodes has been developed to enhance the power output and stability of BMFC systems (Karra
74 et al., 2013; Liu et al., 2015). By integrating multiple pairs of anodes and cathodes with a power
75 management system (PMS) designed with multiple charge pumps, total power output of BMFC systems
76 has substantially increased (Liu et al., 2015).

77 Another problem of BMFC systems is the scalability of anode materials. Dense and compact
78 ocean sediment makes the deployment of anodes a big challenge and exerts a high requirement for the
79 mechanic strength and durability of anodes. Several novel anode materials have been developed to
80 enhance bacterial adhesion and electron transfer by surface modifications (e.g. ruthenium oxide-coated
81 carbon felt, graphene–nickel foam, and graphene–sponges) (Lv et al., 2012; Xie et al., 2012; Wang et al.,
82 2013). High power density (1.6-3.1 W.m⁻²) was achieved in lab tests using pure cultures and enrichment

83 solutions. However, these anode materials have low mechanical strength and fragile structures, which
84 make them unsuitable for directly insertion into sediment. Therefore, it is critical to develop
85 durable/robust anode materials and make them deployable in sediment.

86 The objective of this study was to achieve high power density of BMFCs using two types of
87 cost-effective, durable anode materials (granular activated charcoal (GAC) and carbon cloth (CC)) in
88 novel geometrical setups (inverted tube and rolled) with multiple anodes/electron collectors, and explore
89 the possibilities of maintaining high power density at scale-up BMFCs. Specifically, there were three
90 tasks in this study. First, two novel geometric setups (inverted tube GAC (termed as IT-GAC) and CC-
91 rolled (termed as CCR)) were developed and compared. The effect of the electron collector numbers
92 (titanium (Ti) rods) on power density was examined. Second, the electron collection efficiency of
93 anodes (multiple Ti rods) and the electron acceptance efficiency of cathodes (multiple pieces of cathodes)
94 were correlated to determine the limiting factors of BMFCs. Third, two CCR anodes were arranged
95 contacting to each other in the sediment. The power density of BMFC systems was compared before and
96 after the compact anode matrix in order to determine whether the anode performance was interfered
97 when multiple anodes were inserted compactly in the sediment to fully utilize the sediment content.

98 2. Materials and methods

99 2.1 Mechanisms of IT-GAC and CCR anodes

100 GAC, a cost-effective carbon material with large surface area, high porosity, high conductivity, and high
101 mechanic strength has been tested in BMFC systems. But the performance deteriorated over time due to
102 the clogging of GAC particles in the sediment (Karra et al., 2014). A semi-chamber anode BMFC
103 system had been deployed with the anode chamber located on the water/sediment surface and the anode
104 immersed in water above sediment (Nielsen et al., 2007). Based on this concept, a unique inverted tube

105 GAC (IT-GAC) anode was developed in this study by filling GAC into a tube and was then vertically
106 inserted into the top layer of sediment (Fig. 1). Because the pores between GAC particles were filled
107 with ocean water, the clogging problem was effectively solved.

108 Another durable anode material is flat carbon cloth (FCC), which has been widely used in MFCs
109 treating wastewater and BMFCs (Santoro et al., 2012, 2013; Hsu et al., 2013, Liu et al., 2014a, 2014b).
110 Due to its soft texture, FCC cannot be vertically inserted into sediment to utilize sediment depth
111 effectively, but only be covered on sediment surface. Increasing FCC footprint exhibited significant
112 drop of BMFC power density based on anode footprint area (Hsu et al., 2013). Two innovative
113 approaches were developed in this study to maximize anode surface area in sediment. First, FCC was
114 rolled as the carbon cloth roll (CCR) anode to enhance anode surface area per sediment footprint.
115 Second, multiple non-corrosive and conductive electron collectors (titanium Ti rods) were inserted into
116 the CCR anode to enhance electron collection capacity and strengthen anode robust structure (Fig. 2), so
117 that the CCR can be easily inserted into the sediment to utilize sediment at both horizontal and vertical
118 directions. Previous studies found that more electron collectors inserted into anodes bed led to high
119 power generation in MFCs treating wastewater (Jiang et al., 2011). Compared with complicated
120 chemically treated anode materials (Lv et al., 2012; Xie et al., 2012; Wang et al., 2013), IT-GAC and
121 CCR anodes can be easily manipulated into small footprint and hold a great potential for maintaining
122 high power density in scaled-up BMFC systems, thus achieving high total power output.

123 2.2 IT-GAC and CCR anode setup

124 To prepare the IT-GAC anodes, GAC particles (GAC-4×8 mesh, particle diameter: 2.4–4.8 mm,
125 <http://www.buyactivatedcharcoal.com/>) were filled into a glass tube (Pyrex 9826, diameter 1.5 cm,
126 length: 11 cm) with a footprint of 1.77 cm². The opening of the tube was wrapped with a carbon mesh to
127 prevent the GAC particles from leaking into sediment. Ti rods (diameter 1.6 mm, highly corrosion-

128 resistance Grade 2) that acted as electron collectors were inserted thoroughly into the GAC bed in the
129 tube (Fig. 1C). The Ti rod number (1, 2, and 3) was examined individually to determine the effect of
130 electron collector number on the power output of BMFC systems (Figs. 1B and 1C). Before being
131 inversely inserted into sediment, the GAC tube was filled with ocean water to facilitate biofilm growth
132 on GAC particles.

133 To prepare the CCR anodes, FCC (Untreated Carbon Cloth, Etek Cloth A, Fuelcellearth Company) was
134 rolled compactly into small footprint (diameter 1.2 cm; footprint 1.16 cm², length: 8 cm) (Fig. 2A).
135 Similar as IT-GAC anodes, 1, 2, and 3 Ti rods were inserted individually into the CCR anodes for tests.
136 Before rolling the FCC, Ti rods were “sewed” into the FCC and glued with brush-on electrical tape
137 (BlueMagic®) (Figs. 2B and 2C). The FCC was then wrapped tightly around the Ti rods to form a rigid
138 CCR, and inserted firmly into the top layer of the sediment (Figs. 2A and 2D).

139 2.3 Batch-mode BMFC system

140 All BMFC tests were conducted in batch-mode plastic buckets (height: 38.1 cm; diameter: 28.6 cm).
141 Real ocean water and sediment were acquired from the Long Island Sound (Avery Point, CT). The
142 heights of sediment and water in the buckets were kept 15 cm and 20 cm throughout the whole test
143 period. The IT-GAC and CCR anodes were inserted individually into the sediment. FCC treated with 40%
144 polytetrafluoroethylene (PTFE) and loaded with 0.5 mg cm⁻² Platinum (Pt) powder was used as the
145 cathode in the water phase (Cheng et al., 2006).

146 Large FCC in CCR anodes is expected to increase the effective surface area for biofilm growth. CCRs
147 made from FCCs with two sizes (large FCC: 30 cm×8 cm; small FCC: 10 cm×8 cm) were examined to
148 determine the effect of anode surface area on power density (Fig. 2). In addition, two connection
149 patterns of anodes and cathodes were compared, with the first one being multiple electron collectors

150 connected with a single cathode (Fig. 2A-2), and the second one being multiple electron collectors
151 connected with multiple cathodes (Fig. 2A-3). To simulate the multiple-anode configuration in scaled-up
152 BMFC systems, two CCR anodes were arranged to contact each other in sediment (Fig. 2A-4). The
153 power density of CCR anodes before and after being contacted was compared in order to determine
154 whether the anode performance was interfered when multiple anodes were inserted compactly in the
155 sediment. Each type of anode setup was examined in duplicate tests.

156 Please insert Figure 1 here.

157 Please insert Figure 2 here.

158

159 2.4. Parameter measurement

160 Power generations of all BMFCs unit were measured via conducting polarization curve one week after
161 the startup as previously described (Liu et al., 2014a). Power curve measurement was conducted in
162 duplicated. Power/current density was calculated based on anode footprint area, and the relative standard
163 deviation (RSD) of the power density/current density values was obtained for each anode tested. Voltage
164 values were acquired using a multimeter when the reading stabilized. The oxygen reduction potential
165 (ORP) of sediment and ocean water was measured using an ORP probe (Orion™ Redox/ORP/Temp
166 Electrodes).

167

168 3. Results and discussion

169 3.1. IT-GAC anodes with multiple electron collectors

170 The power generation of IT-GAC anodes was compared in terms of electron collector number. The
171 highest power densities for 1, 2, and 3 electron collectors (Ti rods) were 0.64, 1.71, and 2.92 W m² (per
172 anode footprint) (Fig. 3), indicating that this novel anode pattern significantly increased the power
173 density in ocean sediment (Karra et al., 2013, 2014) and more collectors led to high power generation.
174 These power densities were acquired at the external resistance ($R_{ext.}$) of 66.4 Ω , indicating that the
175 internal resistances (R_{in}) were around 66.4 Ω . Because the R_{in} of a MFC is mainly affected by the
176 resistances of electrolyte and electrodes (Liang et al., 2007; Fan et al., 2008), the low R_{in} of IT-GAC
177 MFCs reflected a good electron passage into the electron collectors that greatly enhanced the power
178 density (Liang et al., 2007). Moreover, the ORP difference between the sediment surface (anode) and
179 the water surface (cathode) was around 500 mV, indicating that the IT-GAC anode based BMFC system
180 is electrochemically feasible for power generation (Tender et al., 2002; Gong et al., 2011). The ORP
181 value found in this study was comparable with a field BMFC test at cold seeps (Nielsen et al., 2008).
182 Previous studies of the inverted semi chamber BMFC (anode brush length: 4 m, coiled inside of an
183 anode chamber on the top of sediment) reached the highest power density of 0.16 W m⁻² (Gong et al.,
184 2011), which was much lower than the power densities of IT-GAC anodes in this study. Small footprint
185 of IT-GAC anodes potentially increased the sediment utilization efficiency per unit sediment footprint.
186 In addition, by inserting multiple Ti rods in IT-GAC anodes, the connection between GAC particles and
187 electron collectors was enhanced and the electron harvest efficiency from GAC particles was
188 significantly improved (Figs. 1 and 3).

189 **Please insert Figure 3 here.**

190 Although the IT-GAC configuration solved the sediment clogging in anodes, there might be two
191 potential problems for long-term operation. First, the discharge of protons (H^+) from the inverted tubes
192 might decline over time. Anaerobic reaction in anodes produces H^+ that normally transfer to cathode and

193 react with oxygen in MFCs (Logan et al., 2006). Because the IT-GAC anode was made by filling GAC
194 into a glass tube, the H^+ produced might be trapped in the tube and accumulated over time, which led to
195 the gradual drop of the BMFC voltage one week after the startup of the IT-GAC anode (data not shown).
196 Previous studies used a valve on the top of the inverted semi-chamber BMFC to discharge the water
197 from chamber to ocean water and prevent the accumulation of protons around anodes (Gong et al., 2011;
198 Nielsen et al., 2007). Thereby, the IT-GAC anodes could be modified with an open end on the top of the
199 tube to release the protons generated in GAC bed. Second, oxygen leakage had been found in the
200 inverted anode chamber due to inhabitation and penetration of aquatic life (e.g. fish and shrimp) in
201 sediment (Nielsen et al., 2007). Even though a mesh was used to prevent GAC loss into sediment (Fig.
202 1B), IT-GAC might be feasible only at a small size to avoid bioturbation.

203

204 3.2. CCR anodes with multiple electron collectors

205 Different number of Ti rods electron collectors (1, 2, and 3) inserted into the CCRs with the same carbon
206 cloth area (FCC size: 10 cm× 8 cm, CCR footprint: 1.2 cm²) was examined. Interestingly, the power
207 density only slightly increased with electron collector number in the CCR anodes (Fig. 4A), with 1.15,
208 1.18, and 1.29 W m⁻² (based on anode footprint area) for 1, 2 and 3 collectors, which were higher than
209 the IT-GAC anode with 1 collector but much lower than those with 2 and 3 collectors (Fig. 3). The
210 possible reason was that electron transfer in CCR anodes was more efficient than IT-GAC anodes. In
211 contrast to CCR that has smooth surface with high conductivity and electrons can be easily transferred
212 within it, IT-GAC anode itself was not well conductive due to the presence of pore water and sediment
213 between GAC particles (Jiang et al., 2011). Single layer of FCC could only be put on sediment
214 horizontally due to its soft structure and incapability of being vertically inserted into sediment (Hsu et al.,
215 2013), while CCR anodes were firm enough to be vertically handled in sediment, which considerably

216 increased the bioavailable surface area per sediment footprint and achieved high power density ($> 1 \text{ W}$
217 m^{-2} anode footprint area, Fig. 4). Unlike GAC anode with the problem of clogging in sediment (Karra et
218 al., 2014), the CCR anodes directly contacted with sediment and greatly facilitated mass transfer.

219 One possible issue for the long term stability of CCR anodes is that the inner layers of CCR might
220 become inaccessible to bacterial cells over time, and the efficiency of the whole multiple-layer CCR
221 structure might decrease. Nevertheless, the CCR anodes made from larger FCC had much higher power
222 density than the ones from smaller FCC (Fig. 4B), demonstrating that the inner layers of CCR anodes
223 were fully utilized for power generation and attached with biofilms. The power density of BMFCs had
224 been found to be positively associated with nutrient contents (e.g. organic carbon and nitrogen) in the
225 sediment pore water that can easily penetrate through multiple CCR layers (Liu et al., 2015), so that
226 biofilms can grow on the inner layers of CCR anodes.

227 **Please insert Figure 4 here.**

228 The power generation of CCR anodes with different FCC sizes (large FCC: 30 cm $\times$ 8 cm, small FCC: 10
229 cm $\times$ 8 cm) rolled into the same CCR footprint area (diameter: 1.2 cm, footprint: 1.16 cm^2) was compared
230 (Fig. 4B). Duplicate tests showed that the CCR anodes made from larger FCC had a much higher power
231 generation (2.55 Wm^{-2}) than those made from small surface FCC (1.49 W m^{-2}) at the R_{ext} of 66.4 Ω .
232 This indicated that at same anode footprint area, more FCCs rolled into the anode will have more surface
233 area available for bacterial adhesion and electron transfer and ultimately lead to high power density. In
234 fact, this is the unique geometric advantage of CCR anode: rolling large FCCs into small footprint (e.g.
235 1-2 cm^2) to achieve large surface area and high sediment utilization efficiency.

236 Much effort has been invested to enhance anode surface properties, accelerate bacterial adhesion and
237 electron transfer efficiency in MFCs. Novel anode materials (e.g. ruthenium oxide-coated carbon felt

(Lv et al., 2012), graphene-coated sponge (Xie et al., 2012), graphene–nickel foam (Wang et al., 2013)) achieved the power density of 1.56~ 3.36 W m⁻² (anode footprint) and 394~ 661 W m⁻³ (anode volume) in pure cultures and phosphorus buffer solution (PBS). However, none of these anode materials were capable of being deployed into real ocean sediment environment due to low mechanical strength and fragile surface property. The power densities (2.92 and 2.55 W m⁻²) of BMFCs with IT-GAC and CCR anodes tested in ocean water and sediment were comparable to those with complicated electrode materials tested in pure cultures and PBS solution (1.56-3.36 W.m⁻²), demonstrating that the anode performance was substantially enhanced by utilizing simple durable anode materials (e.g. GAC and carbon cloth) in novel anode setups (e.g. inverted tube and rolled configurations) and adopting multiple electron collectors.

3.3. CCR anodes with multiple electron collectors being connected with multiple cathodes

Multiple electron collectors (Ti rods) were inserted into an anode and connected with a cathode to enhance power output of a BMFC system (as shown in Figs. 1 and 2). To optimize the power output, three electron collectors of a CCR anode were individually connected with three cathodes (0.5 mg cm⁻² Pt loading) via three R_{ext} (Fig. 2A-3). Connecting multiple cathodes substantially enhanced the electron transfer rate across the biofilms of anodes, which was reflected with enhanced total voltage/current (Table 1). This implied that when electrons are effectively collected from anodes, the electron acceptance capacity of cathode will become the limiting factor. However, most of the BMFC studies reported so far have used single large anode or multi-anode with single collector (Reimers et al., 2001, 2006; Tender et al., 2002; Nielsen et al., 2007; He et al., 2007; Donovan et al., 2008; Nielsen et al., 2008; Tender et al., 2008; Chadwick et al., 2010; Gong et al., 2011; Hsu et al., 2013), in which the electron collection capacity is constrained and leads to the well-known fact that the anode is the limiting factor for BMFCs (Dewan et al., 2008, 2010; Hsu et al., 2013; Ewing et al., 2014). Inserting more collectors

(Ti rods) in anodes can substantially enhance the electron collection/transfer efficiency indicated by increased power output (Fig. 3), and anode may no longer be the limiting factor. Instead, cathodes may become the limiting factor.

Previous study found that scaling up FCC anode based BMFC/MFC systems resulted in a sharp drop in current/power density (Dewan et al., 2008, 2010; Hsu et al., 2013; Ewing et al., 2014), primarily due to the voltage drop across the large anode (internal loss counts for ~30% decrease in the MFC performance). Comparing with anodic electron transfer conducted by electrogenic bacteria, slow oxygen reduction reaction (ORR) rate on cathode could limit the power density of scaled-up MFCs (Logan 2009). The results in this study demonstrated that increasing the number of electron collectors and cathodes could solve the low power density issue of scaled-up MFCs. In order to harvest the electric power of multiple external circuits connected in parallel (Fig. 2A-3), MFCs can be integrated with multiple charging-pump power management systems (PMS) specifically designed for multiple anodes/cathodes (Liu et al., 2015). Thus, individual voltage/power source can be collected to obtain high voltage/power (e.g. 3.3~15 V) capable of supporting sensors.

275

Please insert Table 1 here.

3.4. Performance of the compacted multiple CCR anodes in sediment

By inserting multiple CCR anodes compactly into sediment, BMFC can utilize the sediment capacity as the electron pool and maximize power generation per sediment footprint area. This compact anode matrix will result in the anodes being adjacent to each other, which might affect the mass transfer and electron transfer patterns among anodes and further affect the power generation of individual anodes. The interaction among adjacent anodes should be elucidated in order to scale up BMFCs with multi-

anode configuration. Two CCR anodes were compacted next to each other in sediment to examine the potential impact (as shown in Fig. 2A-4), with each CCR anode (3 electron collectors) being connected with one cathode via a R_{ext} of 325 Ω . Interestingly, the voltage outputs of these two CCR anodes slightly increased (Table 2), implying that the contact of two anodes (large anodic surface area for electrogenic bacteria) might enhance total electron transfer efficiency. The compact multiple-anode configuration did not destabilize the performance of BMFCs, and high power density was maintained by increasing anode number, which paved the way for significantly enhancing power output in scaled-up multi-anode BMFC systems.

Please insert Table 2 here.

3.5. Cost-effectiveness comparison of anode materials

A comprehensive cost-effectiveness comparison of three carbon-based anode materials used in this study (IT-GAC, FCC, and CCR) and two chemically synthetic anodes (ruthenium oxide-coated carbon felt and 3D graphene-nickel foam) developed in other studies was performed based on a sediment area of 1 m² with the assumption that anodes fully occupy the sediment surface (Supporting Information). FCC could only occupy sediment horizontally without penetrating into the sediment depth, while IT-GAC and CCR could utilize sediment both horizontally and vertically. Total estimated cost of multi-CCR anode (\$61036) was about 200 times as single FCC anode (\$295), due to the requirement of more carbon cloth for CCR, while the maximum power output of multi-CCR (2250 mW) was 183 times as that of single FCC (14 mW) (Supporting Information), indicating that the power output of CCR anodes almost linearly increased with the size of carbon cloth rolled (Table S1). CCR is capable of maintaining high power density (>1-2W/m²) in the scaled-up BMFC systems and can achieve high power output, while the power density of FCC rapidly drops when scaled-up (<0.05W/m²) and can't meet the requirement of power output. The estimated cost of IT-GAC (\$887) was lower than CCR, and

306 maximum power output (2920 mW) was similar than that of CCR (Table S1, Supporting Information),
307 however, IT-GAC had the long-term stability problems of bioturbation and proton (H⁺) accumulation.
308 This comparison clearly demonstrated that deploying multiple CCR anodes with multiple electron
309 collectors is economically feasible for high bioavailable surface area, efficient utilization of sediment,
310 and high power output.

311 Chemical synthetic anodes (ruthenium oxide-coated carbon felt and 3D graphene-nickel foam) (Lv et al.,
312 2012; Xie et al., 2012; Wang et al., 2013) required complicated fabrication processes (e.g.
313 electrodeposition, thermal treatment) and expensive chemicals (e.g. ruthenium, nickel, precursors)
314 (Table S1), leading to a much higher cost than the carbon-based anodes with simple fabrication
315 processes (FCC, GAC, and CCR). The power output of these chemical synthetic anodes was similar as
316 that of simple durable anode materials used in this study. Moreover, these anode materials had been
317 examined only in pure culture and/or PBS solution due to fragile structure and weak mechanic strength.
318 Long-term sediment deployment is questionable for these chemically synthetic anodes.

319 **3.6. Significance of achieving high power density in BMFCs using multiple electron collectors**

320 Sediment in ocean environment has high density and compact structure due to high ocean water pressure.
321 To effectively harvest the electric power from sediment using BMFCs, one strategy is to roll large FCCs
322 into small footprint for bacterial adhesion and electron transfer, and increase anode mechanic strength
323 (FCC wrapped around rigid Ti rods) for feasible deployment in sediment. The BMFC systems with the
324 IT-GAC anodes and CCR anodes had the power density of 2.92 and 2.55 W m⁻², which were the highest
325 power density ever reported in BMFCs, and comparable with the complicated chemically treated anode
326 materials tested in pure cultures and PBS solutions. Especially, the CCR anodes well utilize the 3-D
327 content of sediment and can be directly inserted into sediment without the problems of clogging and
328 fragility. High power density and stable performance of the compacted multiple CCR anodes proved that

organizing multiple anodes adjacent to each other is feasible to maximize the utilization of sediment and maintain high total power output with increased anode footprint.

Until now, scaling up MFC systems has been achieved by increasing the size and/or volume of the anodes/cathodes. However, due to resistive losses of electrodes and sediment, the power density dropped in magnitude orders at the scale-up BMFCs. In order to fully utilize sediment electron pool and minimize electron transfer resistance, deploying multiple small footprint anodes in sediment is the most feasible approach so far. These findings underlined the importance of optimizing electrodes at small footprint for scale-up BMFCs.

4. Conclusions

Two types of novel anode configurations using cost-effective durable electrode materials (IT-GAC and CCR) were developed to achieve high power output of BMFCs. The batch-mode lab tests using ocean water and sediment showed that the IT-GAC and CCR anodes equipped with 3 electron collectors achieved the highest power densities of 2.92 and 2.55 W m⁻². More electron collectors in an anode substantially enhanced the power density. Two CCR anodes adjacent to each other did not show the interference of power generation, indicating that multiple small anodes can be inserted compactly in sediment to fully utilize the sediment content and maximize the power density per sediment footprint. The results revealed an innovative approach of maintaining high power density (> 1 W/m² sediment footprint) in scaled-up BMFC systems by using multiple-anode structures and cost-effective durable electrode materials with multiple electron collectors.

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Table 1 Performances of Single CCR anode connected with multiple cathodes via multiple electron collectors (R_{ext} . 325 Ω)					
Connecting with 1 cathode		Connecting with 3 cathodes			
		With cathode 1#	With cathode 2#	With cathode 3#	Combining 3 cathodes
Voltage (mV)	185.7 $\pm$ 4.5	145.8 $\pm$ 5.0	163.6 $\pm$ 15.7	150.7 $\pm$ 16.5	460.1 $\pm$ 37.2
Current (mA)	0.57 $\pm$ 0.014	0.45 $\pm$ 0.015	0.5 $\pm$ 0.048	0.46 $\pm$ 0.051	1.42 $\pm$ 0.11
Current density (A m ⁻²)	5.71 $\pm$ 0.14	4.48 $\pm$ 0.15	5.03 $\pm$ 0.48	4.63 $\pm$ 0.51	N/A
Power density (W m ⁻²)	1.06 $\pm$ 0.05	0.65 $\pm$ 0.05	0.83 $\pm$ 0.16	0.71 $\pm$ 0.15	N/A

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Table 2 Performances of two CCR anodes before and after being contacted in sediment (R_{ext} 325 Ω)				
		Voltage (mV)	Current density ($A\ m^{-2}$)	Power density ($W\ m^{-2}$)
Before two anodes contacted	Anode #1	190.2	5.85	1.11
	Anode #2	181.2	5.56	1.01
After two anodes contacted	Anode #1	216.6	6.66	1.44
	Anode #2	201.7	6.21	1.25

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417 **Fig. 1** (A) Schematic of IT-GAC anodes based BMFC; (B) the top view of multiple Ti rods (as electron
418 collectors) inserted into the GAC tube; (C) the image of IT-GAC anodes with multi electron collectors.

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Fig. 2 (A) Schematic of CCR anode based BMFCs; (B) the top view of multiple Ti rods (as electron collectors) inserted into the CCR anode; (C) CCR anodes made from FCC with multiple electron collectors (Ti rods); (D) the assembled CCR anodes with 1, 2, and 3 electrode collectors (Ti rods).

Fig. 3 Power density (W/m^2) of the IT-GAC anodes with 1, 2, and 3 electron collectors. (The RSD of power density for the IT-GAC anodes with 1, 2, and 3 electron collectors were 0.084, 0.128, and 0.080, respectively. The RSD of current density for the IT-GAC anodes with 1, 2, and 3 electron collectors were 0.094, 0.079, and 0.073, respectively)

Fig. 4 Power generations of CCR anodes with different electron collectors and FCC sizes. (A: 1, 2, and 3 electron collectors in CCR anodes with the same FCC size (10 cm×8 cm). The RSD of power density for CCR anodes with 1, 2, and 3 electron collectors were 0.061, 0.076, and 0.068, respectively; the RSD of current density for CCR anodes were 0.061, 0.071, and 0.070, respectively). B: CCR anodes with 1

438 electron collector made from two FCC sizes (big FCC: 10 cm×8 cm; small FCC: 30 cm×8 cm). The
439 RSD of power density for two big CCR anodes (1 and 2) and two small CCR anodes (1 and 2) were
440 0.068, 0.084, 0.071, and 0.068, respectively; the RSD of current density of CCR anodes were 0.077,
441 0.090, 0.075, and 0.066, respectively).