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Power from marine sediment fuel cells: the influence of anode material

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Abstract The effect of anode material on the performance of microbial fuel cells (MFC), which utilise oxidisable carbon compounds and other components present in sediments on ocean floors, estuaries and other similar environments is reported. The MFC anode materials were carbon sponge, carbon cloth, carbon fibre, and reticulated vitreous carbon (RVC). Power was produced through the microbial activity at the anode in conjunction with, principally, oxygen reduction at a graphite cloth cathode. After a period of stabilisation, open circuit voltages up to 700 mV were observed for most cells. Steady state polarisations gave maximum power densities of 55 mW m^{-2} using carbon sponge as the anode; which was nearly twice that achieved with carbon cloth. The latter material typically gave power densities of around 20 mW m^{-2}. The performance of the cell was reduced by operation at a low temperature of 5 °C. Generally, for cells which were capable of generating power at current densities of 100 mA m^{-2} and greater, mass transport was found to limit both the anode and the cathode performance, due primarily to the low concentrations of electro-active species present or generated in cells.

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1 Introduction

An abundant supply of fuel is available in marine sediments, in the form of oxidisable organic carbon. Sediments containing around 2 wt% organic material have energy density capabilities of approximately 17 Wh dm^{-3}. Micro-organisms limited by oxygen supplied by overlying sea water can create potential differences of up to 0.8 V over a few millimetres of sediment. With such vast supplies of fuel, possibilities abound for applications requiring low levels of power. One example is to generate power in oceanographic research. With the aid of microbial fuel cells, various monitors can be powered, e.g., temperature, salinity, tidal patterns, the presence of algae and other life forms, migration patterns of fish and other marine wildlife, organic contamination from oil production and metallic compounds from other industrial processes. Powering such sensors using fuel cells is potentially a better long term option than using batteries. The batteries that currently provide the power for oceanographic instruments need to be changed periodically and with this brings obvious costs. Sustained power; of 30 mW m^{-2} for over 1.5 years has been demonstrated from microbial fuel cells operating in marine environments [1].

Redox processes in the ocean are fuelled by organic matter as the ultimate reductant and oxygen as the ultimate oxidant. Energy can be harvested from aquatic sediments by burying an electrode (the anode) in anoxic sediments and electrically connecting to a second electrode (the cathode) in the overlying aerobic water [1–4]. Biological fuel cells use the natural catalytic ability of microorganisms to oxidize a wide variety of substrates, while producing electrons in a form that can be harvested at an electrode. For example, fuel cells utilising sulphate-reducing bacteria have been constructed, with the microbially produced hydrogen sulfide serving to shuttle electrons to the electrode surface [5, 6]. In addition, Fe(III)-reducing microorganisms such as *Shewanella putrefaciens* [7, 8], *Clostridium* [9], *Aeromonas hydrophila* [10], *Rhodoferax ferrireducens* [11], *Desulfobulbus propionicus* [12], and several species of *Geobacteraceae* [1, 13] have been shown to directly transfer electrons from the oxidation of organic compounds to the surface of an electrode without the need for an external mediator.

In sediment fuel cells, natural populations of microorganisms in the anoxic sediments are responsible for electron transfer to the current-harvesting anode. Previous culture-independent studies and studies of organisms cultivated from fuel cell anodes have indicated that a specific group of Fe(III)-reducing microorganisms, the *Geobacteraceae*, is primarily responsible for electricity production in some sediment fuel cells [1, 2, 4]. Further analysis of pure cultures of *Geobacteraceae* has shown that these organisms can conserve energy to support growth by oxidizing organic compounds to carbon dioxide, with an electrode serving as the sole electron acceptor [1, 2]. Therefore, a likely explanation for the functioning of sediment fuel cells is that microorganisms in the sediments convert the complex sediment organic matter to fermentation products, most notably acetate, and that *Geobacteraceae* colonizing the anode oxidize these fermentation products to carbon dioxide, with transfer of electrons to the current-harvesting electrode [1]. These electrons then flow to the cathode in the overlying aerobic water, where they react with oxygen [3].

A reaction mechanism in the marine sediment fuel cell has been proposed in which a series of microbial reductions take place progressively in the surface sediment layer, forming water from oxygen, and Mn^{2+}, Fe^{2+}, S^{2-} from MnO_2, Fe_2O_3 and SO_4^{2-} respectively [14]. These reactions provide the open circuit potential of the cell and promote oxidation of sediment organic matter, $(CH_2O)_6$. Reactions that occur at the anode include oxidation of sulphide to sulphur, and oxidation of acetate, to CO_2, and sulphur, to SO_4^{2-}, (and potentially Fe^{2+} to Fe_2O_3) by micro-organisms colonising the anode [1, 2, 3]. The products of these oxidations can again be microbially reduced by bacteria. At the cathode, dissolved oxygen in the seawater is reduced.

The net reaction is oxidation of organic matter by dissolved oxygen catalyzed by sediment-dwelling anaerobes and mediated by the intermediate oxidants reduced in anaerobic respiratory reactions, e.g.:

$$(CH_2O)_6 + 6\,O_2 \rightarrow 6\,CO_2 + 6\,H_2O \tag{1}$$

The consequent depletion of oxygen in the sediment provides enough separation between oxygen and organic matter to provide an external circuit for electron transfer.

In order to rationally optimise the harvesting of electricity from aquatic sediments, it is necessary to understand the mechanisms of microbe-electrode electron transfer. Ideally, this phenomenon should be studied in microorganisms known to colonise energy-harvesting electrodes in sediments. Holmes et al. [12] reported two strains isolated from the surfaces of electricity-harvesting electrodes incubated in marine sediments. Both isolates represent a novel genus in the *Geobacteraceae* with the capacity to grow at temperatures (4 °C) lower than those previously reported for microorganisms in this family [15]. These organisms can also oxidise a variety of electron donors, with electrodes and Fe(III) serving as the terminal electron acceptors. This was the first report of microorganisms isolated from electrode surfaces that can effectively oxidise organic compounds to carbon dioxide with an electrode serving as the electron acceptor.

In previous studies of sediment fuel cells Tender et al. [4] (2002) have created a simple microbial fuel cell using different sediments on the sea floor, based on two carbon electrodes placed in two different environments: one in the anoxic sediments and the other in the seawater immediately above the sediment. The peak power density of the sediment fuel cell was around 30 mW m^{-2}, at a current density of 75 mA m^{-2} and cell voltage of 400 mV. Recent work [14] has shown that by using modified anodes incorporating anthraquinone, quinine and manganese and iron oxide species an increase in power density, up to approximately 100 mW m^{-2} can be achieved in a marine sediment cell. However this power was not maintained over extended periods and declined gradually with time to levels obtained with unmodified anodes.

Several groups have investigated anode materials in microbial fuels to try to increase the power output per unit volume of reactor. Logan et al. [15] investigated graphite brush anodes, consisting of graphite fibres wound around metal core. Using a simulated wastewater MFCs gave up to 1430 mW m^{-2} with brush electrodes, compared with 600 mW m^{-2} with a plain carbon paper electrode.

A carbon nanotube (CNT)/polyaniline (PANI) composite has been evaluated as anode material for high-power microbial fuel cells [16]. The maximum power density obtained was 42 mW m^{-2} with Escherichia coli as the microbial catalyst, which was significantly greater than that achieved with polyaniline alone.

Ammonia gas treatment of a carbon cloth anode has been used to increase the surface charge of the electrode and improved MFC performance [17]. Power increased to nearly 2000 mW m^{-2} using a phosphate buffer with the ammonia-treated electrode.

The purpose of this study was to investigate the influence of different un-modified electrode material on the performance of marine sediment microbial fuel cells producing low levels of power. The materials used were restricted to a selection of carbon materials with relatively high surface areas and which were not chemically modified; in an attempt to maintain long term operation.

2 Experimental

2.1 Microbial fuel cell

Figure 1 shows a schematic of the MFC, which consisted of an inert cylindrical container, 16 cm in diameter and 16 cm high. The anode was 11 cm in diameter, and was buried 3 cm below and parallel to the surface of the sediment. The cathode, 15 cm in diameter, was positioned in the seawater, 2–3 cm above and parallel to the sediment surface. The seawater layer above the cathode was 5 cm in depth. No separator was used in the cell. Air supply to the cathode was thus by natural diffusion from the surface of the seawater unless otherwise stated.

The anode and cathode were supported by plastic rings to maintain their horizontal positions. The fuel cells were maintained at room temperature except when the effect of temperature was investigated. Cells using different anodes were tested, in duplicate and simultaneously, using the same sources of sediment and seawater, over the period of the study, which for some cells was 9 months.

Silver chloride references electrodes were connected to the cells, via luggin capillary probes, to measure electrode potentials. Current densities quoted in this paper are based on the anode cross sectional area.

The marine sediment and seawater were collected from the North Sea, off the north-east coast of England (55°01′08″N, 1°25′11″W) at a depth of 62 m. The materials were transported under reduced temperature to restrict microbial growth, to nearby laboratory facilities

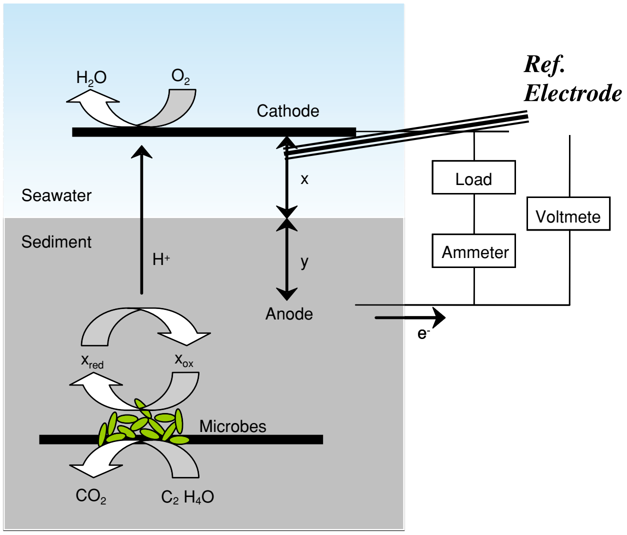

Fig. 1 Schematic representation of the sediment microbial fuel cell

As part of the test protocol, the cells were initially left to equilibrate, while monitoring the open circuit voltage (OCV). When the OCV stabilised, an external circuit was established, with electrodes connected through a variable resistor load, using either a DC power supply or a variable resistor. The variable resistor was used to generate cell current density and voltage polarization data. Current generated across the cell, of the order of microamps, was measured using a multimeter, while continually monitoring the voltage via a second multimeter. Other parameters monitored were, electrode potentials, pH, and dissolved oxygen (YSI 550A dissolved oxygen meter) in the water and in the sediment.

2.1.1 Electrodes

Anodes used in this study were all carbon and were selected because of their different characteristics (porosity, morphology, carbon type). The materials used were carbon cloth (Avcarb 1209 HC, Ballard), carbon paper (Toray Carbon Paper TGP60 0WP), carbon sponge, graphite (5 mm thick disk) and reticulated vitreous carbon (RVC). The RVC anode was 1.2 cm thick, 100 ppi (nominal pores per linear inch) grade. RVC is a porous, rigid material with an open structure and good electrical conductivity. Electrodes were pre-treated before use, by washing in acetone for 15 min, followed by boiling in 1.0 mol dm^{-3} (M) HCl for 15 min and then rinsing in de-ionised water.

The cathode used in all cells was carbon cloth. Silver chloride references electrodes connected to the cathodes using luggin capillary probes were used to measure electrode potentials.

3 Results and discussion

3.1 Performance of the sediment fuel cell

At the beginning of the tests, the fuel cells were left to run at open circuit to allow bacterial colonisation of the anode and cathode. The voltage of each MFC was monitored daily, until it reached equilibrium, which suggested that the anode had been colonized by a “stable” bacterial community. The MFCs needed several weeks to equilibrate until the anodes were colonised by the bacterial population from the marine sediment. The equilibration period of the cells, i.e. the time to reach a relatively stable potential, varied with the electrode materials used; this fact can be attributed to the different morphologies, structure and surface characteristics of the electrode materials and potential “biocompatibility” with the bacterial populations

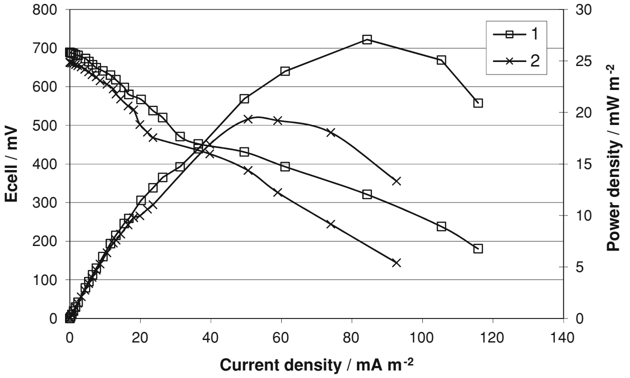

Fig. 2 Cell voltage and power density curves of the sediment fuel cell with carbon cloth anodes. Two sets of data are for polarisation in successive weeks of the study. □: first polarisation; ×: second polarisation

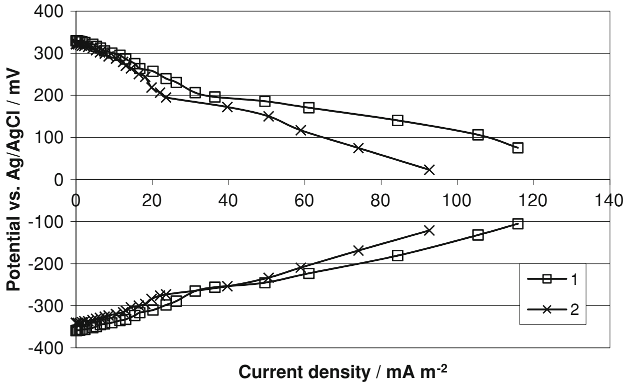

Fig. 3 Anode and cathode polarisations for cell with a carbon cloth anode. □: first polarisation; ×: second polarisation. Upper curve: cathode polarisation. Lower curve: anode polarisation

in the sediment and seawater. The OCPs gradually rose over the initial days and weeks of operation. Fluctuations in voltage arose, presumably because cell temperatures were not controlled, but varied according to the night and day time temperature of the laboratory and the fact that the cell relied on natural diffusion of oxygen from the water surface.

For the cells with carbon cloth electrodes, open circuit potentials were 650–700 mV (Fig. 2). The peak power densities were 20–27 mW m^{-2}, at cell voltages around 350 mV, and peak current densities of 60 to 90 mA m^{-2} respectively (Fig. 2). The data reflect the range of performance that was obtained from any single sediment cell tested over a period of months. The power densities are similar to those obtained by Tender et al. [4], i.e. 17–30 mW m^{-2} using graphite electrodes.

The dissolved oxygen (DO) concentration in the sea water, at 100% saturation with air, was measured at 9.2 mg dm^{-3}. The DO levels in both sediment and water fell gradually in the first 3–5 days and then rose to almost constant values of 83% and 56% of saturation for water and sediment respectively. Although these measurements were made away from the electrodes they do indicate that the MFCs were operating under partially aerobic conditions. Variations in pH were small, typically rising from around 7.6 to 8.0 over the duration of the tests.

Figure 3 shows the variation in anode and cathode potentials with current density for cells with carbon cloth electrodes measured at two separate times (three days apart). The anode polarisations were approximately the same, although the cathode polarisations were somewhat different; the second polarisation started at a lower OCP of 660 mV. The fall in potential for the second polarisation was greater than that for the first and the difference between the potentials, at a fixed current density, increased as the current density increased. This difference in potential is essentially mapped in the cell voltage characteristics shown in Fig. 2.

One cell with a carbon cloth anode was operated at 5.0 °C; a temperature close to that experienced in the marine environment. The open circuit potential started at 500 mV which was lower than that obtained at room temperature. After two polarisations the peak power density was 15.0 mW m^{-2}; smaller than that achieved at room temperature (Fig. 2), although clearly showing that reasonable power can be obtained at lower temperatures.

The anode and cathode polarisations of the MFC operated at 5 °C were also measured. At low overpotentials cathode polarisation characteristics were linear. On increasing load, the current density rose approximately exponentially with overpotential, until mass transport limitations appeared [18]. Measured dissolved oxygen concentrations were typically 7.5 mg dm^{-3}, i.e. 2.5 mmol dm^{-3} (mM) [19]. At this concentration mass transport limiting current densities, j_l, for oxygen reduction are approximately

$$j_l = n\,F\,k_l C_{O_2} = 10^9\,k_l \quad \mathrm{mA\,m^{-2}} \qquad (2)$$

where C_{O_2} is the dissolved oxygen concentration, F is Faradays constant, n is the number of electrons in the reduction of oxygen (4) and k_l is the mass transport coefficient (m s^{-1}) for oxygen transport to the electrode in an essentially stationary fluid.

With a mass transport coefficient, for a horizontal electrode in a stationary fluid, of the order of 10^{-7} (m s^{-1}) [20], the limiting current density would be around 100 mA m^{-2}. This value is in agreement with apparent limiting current behaviour observed experimentally.

The extent of cathode polarisation (change in potential) was greater than that of the anode polarisation, e.g. 80 mV cf. 40 mV at 40 mA m^{-2}. Comparing data for the two operating temperatures, the cathode had a significant influence on cell behaviour, with greater polarisation at 5 °C. This would indicate that the potential impact of slower reaction kinetics at the lower temperature was only partially counteracted by a greater oxygen concentration at the lower temperature. Overall the anode was not the limiting factor on cell performance and the microbial community functioned effectively at the lower temperature.

The anode polarisation characteristics were somewhat similar to that of the cathode although the extent of polarisation was lower. However, it is not possible to say that the increased anode overpotential, at higher current densities, was due to mass transport limitations or to change in reactions and mechanisms for microbial based reaction. Many factors would be influencing the anode behaviour including direct and indirect electron transfer at the anode, metabolic kinetics, mass transfer of mediators and mss transfer inside then biofilm.

3.2 Comparison of anode materials

Average cell potential and power density vs. current density data; for cells using carbon sponge, carbon fibre and RVC anodes, was taken over a period of 18 days. The carbon sponge anode gave power densities between 51 to 54 mW m^{-2} at cell voltages of 300 mV and current densities of around 170 mA m^{-2}. The carbon fibre anode gave a peak power density of 4.5 mW m^{-2} and the RVC anode gave a low, peak power density of 0.18 mW m^{-2} at a peak current density of 1.0 mA m^{-2}. The poor cell performance with the RVC anode was due mainly to the relatively large anode overpotential, e.g. 150 mV at a current density of 1.0 mA cm^{-2}.

Figure 4 shows the anode and cathode polarisations of the carbon sponge, carbon fibre and RVC based cells; obtained during the cell polarisations. The cathode polarisations were similar for each cell; due to the fact that they all had a carbon cloth cathode. The anode clearly had a more significant influence on cell performance than the cathode. For example, at an anode overpotential of 100 mV (measured from the potential at zero current), the current density with carbon sponge was around 100 mA m^{-2}, compared with only 50 (see Fig. 3), 5 and 0.8 mA m^{-2} for carbon cloth, carbon fibre and RVC respectively.

Although microbial electrochemical kinetics are complex, the polarisation behaviour of the anodes (particularly carbon fibre and RVC) exhibited characteristics somewhat typical of simple electrode reaction. At low overpotentials the current density increased almost linearly with an increase in potential. At higher overpotentials, current density rose more rapidly; partly as a result of the characteristic rise in current density with overpotential for electrochemical reactions; although the variation in current density was undoubtedly influenced by other factors such as mass transport of substrate, mediators etc.. Anode potentials were not IR corrected and were determined indirectly from the difference in cell potential and cathode potential.

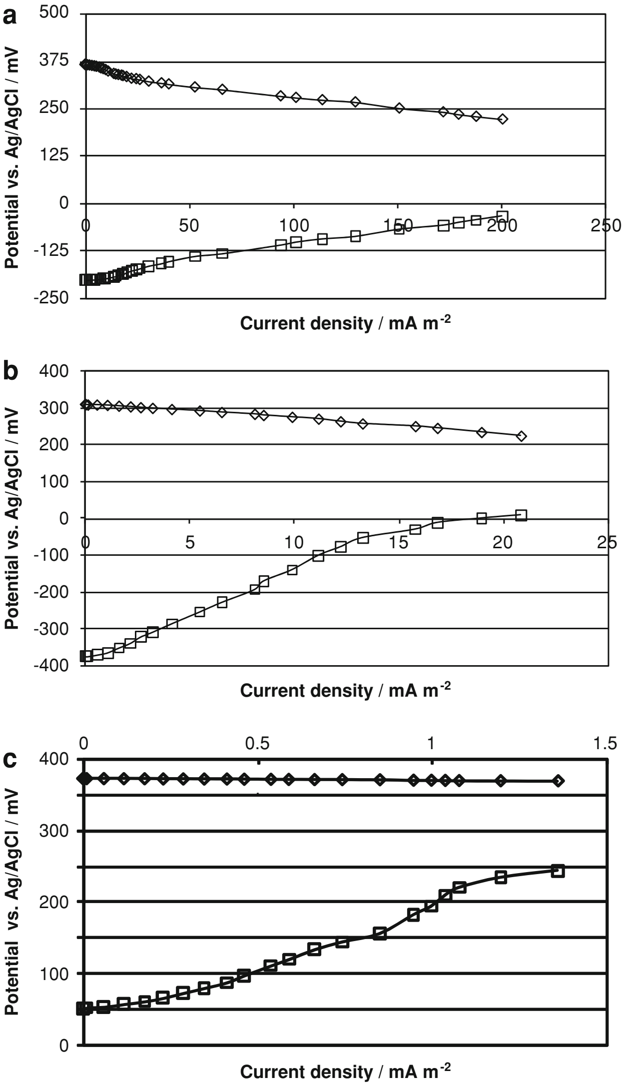

Fig. 4 Anode and cathode polarisation curves for cells with different carbon anodes. Room temperature. Carbon cloth cathode. (**a**) carbon sponge, (**b**) carbon fibre (**c**) RVC. Upper curve (◇): cathode polarisation. Lower curve (□): anode polarisation

Previous researchers have characterised anode polarisation using Tafel plots of anode overpotential, in the range 60–100 mV [14]. In this study [14], using linear sweep voltammetry (at 1.0 mV s^{-1}) with graphite and “catalysed” or doped graphite electrodes, the relative activity of the anodes was compared by obtaining apparent exchange current densities from the intercepts of log (j) vs. overpotential plots (at zero overpotential measured against the open circuit potential). The slopes of the Tafel plots were typically 250 mV per decade [14].

In the case of the data from our study, which were obtained during cell voltage measurements under variable load, an approximate exponential rise in current density with overpotential can be seen (Fig. 4), except for carbon sponge. In the case of carbon sponge the variation of current density with overpotential was probably influenced by internal resistance (IR) voltage loss and the onset of some mass transport limitation, at the higher current densities experienced with this anode. For carbon sponge, the change in overpotential (Tafel slope) in the lower overpotential range, of 60–100 mV, was approximately 100 mV per decade (Fig. 4a). This would suggest that the carbon sponge anode had more favourable electrochemical characteristics compared to graphite; although the latter material used by Lowy et al. [14] was not porous.

To compare the relative activity of the anodes in this work the electrochemical behaviour was quantified by using the low overpotential range (<40 mV), linear approximation of the Butler Volmer equation

$$j = 2aj_oC\alpha nF\eta/RT = 2aj_oC\eta/b \qquad (3)$$

where, j_o, is the exchange current density, α, is the transfer coefficient, a, is the active area per unit cross sectional area of the electrode and depends on the particular material used. C is the concentration of active species and $b = RT/\alpha nF$ is the Tafel slope.

Thus from (1), a plot of overpotential against current density (or specifically $j/2$) gives a slope, b/aCj_o (ohm cm^2), a measure of charge transfer resistivity. Low Tafel slopes and high exchange current densities thus give lower charge transfer resistivities and are a measure of relative anode activity.

The values of overpotential were measured against the mixed potential for a particular anode material measured at zero current density. Table 1 compares the slopes of the plots of overpotential against current density, i.e. kinetic resistivity, for the anodes tested in this work. The resistivities vary from 1.0 (sponge) to 100 (RVC) ohm cm^2 and the relative activity of the anodes corresponded to their relative performance in fuel cells.

Table 1 also compares the performance (the peak power density) of the fuel cells used in this study. The data clearly identify carbon sponge as a potential electrode material for the sediment fuel cell giving peak power densities above 50 mW cm^{-2}. This performance is approximately 2.5 times that reported by Tender [4] for graphite anodes, although it is approximately 55–60 % of that reported by Lowy et al. [14] for modified graphite anodes incorporating anthraquinone disulphonic acid (AQDS) or Ni^{2+} and Mn^{2+}. However, in the case of the latter modified anodes, power performance decayed with time and fell to values achieved with the un-modified graphite anode (~20 mW m^{-2}) [15].

Table 1 Peak power densities and electrochemical activities of MFC with different carbon anodes

Anode	Peak power density (mW m^{-2})	Anode charge transfer resistivity/ohm cm^2 (b/aCj_o)
Carbon sponge	55	1.0
Carbon cloth	19–27.5	3.0
Carbon cloth (5 °C)	15	3.4
Carbon fibre	4.5	20
RVC	0.2	100

4 Constant load testing

The MFCs used in this study were maintained under a constant load (380 Ω resistor), which gave potentials close to the point where the peak power density was obtained, and continuously monitored to study their stability over 329 h. Typical data are shown in Fig. 5 for the MFC with the carbon cloth anode. The potential increased from approximately 125 mV to 250 mV during the first 100 h, after which the potential was relatively constant; with small variations with time between 200 mV and 300 mV (Fig. 5). The variation in potential in the approximate steady state period can be attributed to the changes in the laboratory temperature from daytime to night time operation- cell temperatures were not controlled.

A set of MFCs was also maintained under a constant current of 0.8 mA (~90 mA m^{-2}) for 530 h and to study their stability. The potential was relatively constant; varying between 0.3 and 0.34 V over the period of the test. Overall when cells were maintained under a constant current, the potentials were relatively constant within the constraints of fluctuations in laboratory temperature during day and night and lack of agitation of the seawater, making it susceptible to external conditions. Ideally a field study in larger tanks containing fresh oxygenated seawater, with a

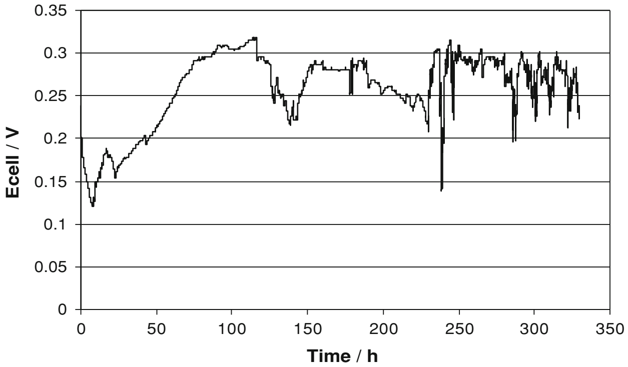

Fig. 5 Variation of MFC voltage with time. Monitoring of cell with carbon cloth anode maintained under a constant resistance

system of agitation to simulate the sea currents should be carried out.

5 Conclusions

Microbial fuel cells which utilise sediments from ocean floors have been demonstrated using different carbon anodes: carbon sponge, carbon cloth, carbon fibre, graphite and reticulated vitreous carbon. After a period of stabilisation, open circuit potentials between 600–700 mV were observed for most cells. Steady state polarisations typically gave maximum peak power densities of 55 mW m^{-2} for carbon sponge anodes, which was more than double that achieved with carbon cloth and graphite. These latter materials typically gave power densities of around 20 mW cm^{-2}. The performance of the cell was reduced by operation at a low temperature of 5 °C. Generally, for cells which generated power at current densities of 100 mA m^{-2} and greater, mass transport was found to limit anode and cathode performance, due primarily to low concentrations of oxygen and electro-active species present or generated in cells. The MFCs gave sustained power under constant load operation.

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