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Cathode materials for ceramic based microbial fuel cells (MFCs)

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ABSTRACT

This study showed the electrochemical performance of different cathode electrodes tested on a ceramic separator functioning as a cation exchange membrane. Particularly, three different carbonaceous-based materials (carbon cloth (CC), carbon mesh (CM) and carbon veil (CV)) have been used as an electrode and as the current collector. When used as an electrode, CC outperformed the others. The carbonaceous materials have been modified using conductive paint (PA) and micro porous layer (MPL). With these modifications, the current output was two–three times higher. Generally, the current produced was slightly higher with MPL treatment compared to PA except in the case of CV-MPL that had lower output probably due to the negative effect of the heat treatment on the mechanical strength of the CV. In the case of PA, the current collectors do not seem to affect the output. The same consideration can also be done for the MPL except for the CV. The surface morphology seems to explain the results. Linear correlation was found between current produced and nanoscale roughness and skewness. The results indicated that those morphological parameters increased the contact between the cathode and the ceramic surface, thus enhancing the current generated. The further addition of the inorganic non-platinum group catalyst (Fe-AAPyr) on the surface significantly enhanced the performances. Following MPL modification and MPL-Fe-AAPyr addition, CM was the most cost effective support. CV was the most cost effective support with PA modification.

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Introduction

Water availability has been an important global challenge and consequently, water treatment is critical in successfully addressing this problem. The common technologies available for wastewater treatment are very efficient in degrading organics, but very expensive to operate, mainly due to the electricity necessary for mixing and pumping oxygen. Alternative approaches, such as Microbial Fuel Cells (MFCs), have been explored to make the process more efficient and to decrease significantly the cost [1,2]. This emerging bio-electrochemical technology is not only able to degrade organics but also to transform the chemical energy stored in compounds such as water pollutants into useful electricity [1,2]. The electricity generated can be used for small-scale real applications [3–5]. However, this technology is still at the lab scale of development.

Major concerns related to MFC utilization are: i) the relative high cost of the electrode materials [6,7]; ii) the low power output caused by the unoptimized anodic [8] and cathodic [9] conversion processes.

Anodic processes are mainly controlled by the electrically active bacteria, which upon adhering to the electrode, can degrade organics releasing and transferring electrons to the conductive electrode. Electrons moving through the external circuit generate electricity that can be harvested and successfully utilized [3–5]. The understanding of bacteria attachment [10], electron transfer [11], biofilm formation and development [12,13] bacteria selection [14] is still a matter of ongoing investigation.

Cathode processes are mainly affected by the high overpotential caused by the low electrochemical activity of the catalysts used (inorganic or biotic) at neutral pH [15]. Although it has been shown [16,17] that specific enzymes can significantly lower the overpotential of oxygen reduction reaction (ORR) at neutral pH, utilization of carbonaceous [6,18] or transition metal-based [19,20] catalysts is preferred due to the higher availability, low cost and durability. Platinum has been traditionally used as a cathodic catalyst in MFCs [21] but it has been shown to suffer from rapid decrease in performance due to the fast poisoning effect of sulfide presence in the wastewater [22]. Two different avenues are currently being exploited aiming at a finding the trade-off between cost and efficiency of ORR: i) catalysts based on utilization of carbon-based materials, having high conductivity and high surface area [6]; ii) inorganic catalysts such as iron (Fe) [23], cobalt (Co) [24,25] and manganese (Mn) [26].

Moreover, the current collector design is also very important for guaranteeing high cathode performances. Several current collectors have been used in MFCs mainly based on carbonaceous materials and particularly carbon veil [27,28], carbon cloth [29], carbon paper [30] and carbon felt [30]. Also, metallic meshes have been used, based on corrosion proof stainless steel [31]. An understanding of the best performing and cost-effective material is still necessary.

Some studies have shown that the formation of biofilm due to the direct exposure of the anodic solution in membraneless MFC configuration lead to an enhancement in the cathode performance as a result of a biocathode formation [32,33]. The

OH⁻ production during the ORR leads to cathode alkalization [34,35] and calcium and sodium carbonate precipitation on the cathode [36], lowering its long-term operation [32]. Consequently, the option of using a solid separator able to decrease the negative effects of cathode alkalization seems to be reasonable for preventing cathode deactivation and keeping the anode chamber under strictly anaerobic conditions. Anionic and cationic exchange membranes have been used previously in a single chamber or double chamber MFCs [37]. It has been shown that cationic membranes are preferable than the anionic membranes most likely because they prevent an accumulation of protons at the anode that inhibits bacterial metabolism [38]. The main problem related with solid polymeric separators is the high cost of the membrane that makes them not suitable for a large-scale operation [39]. Recently, MFCs with ceramic cation exchange membranes, utilized as a physical separator between the anode and cathode have been successfully developed and explored [39,40]. The main advantages of ceramic separators are: i) low cost; ii) high ions selectivity; iii) high mechanical strength and iv) high durability [39,40].

This work focuses on the electrochemical analysis of different low-cost carbonaceous materials suitable for the design of cathodes in ceramic MFCs. Carbonaceous materials (veil, cloth and mesh) have been tested: i) without any pre-treatment, ii) coated with a micro porous layer (MPL); iii) coated with a conductive carbon paint. The performance of low-cost non-platinum group metals (non-PGM) Fe-Aminoantipyrine (Fe-AAPyr) has also been studied, demonstrating promising results compared to other materials. Cost-performance analysis of the different options has been also carried out, in the light of future large-scale applications.

Materials and method

Cathode materials

Different materials have been tested as cathodes or cathodic support in ceramic-based MFC. Particularly, three carbonaceous electron collectors have been used identified as carbon veil (CV), carbon cloth (CC) and carbon mesh (CM) (Fig. 1). Those materials were purchased from PRF Composite Materials (Dorset, UK), Saati (Legnano, Italy) and Electromar (Milan, Italy), respectively. CV, CC, and CM have been used as controls during the experiments.

Modifications were done with the addition of conductive paint from TIMCAL Ltd. Switzerland (PA) or a micro porous layer (MPL). The materials with PA and MPL have been additionally modified with Fe-AAPyr as a catalyst for ORR. PA was applied on the carbonaceous support using a brush, covering the entire surface with a PA loading of $40 \pm 10 \text{ mg cm}^{-2}$. MPL was done similarly as previously described [41]. In summary, 0.7 g of TIMCAL carbon powder was put in a beaker with 9.1 mL of distilled water and 21.5 mL of nonionic surfactant (Triton X100, Sigma Aldrich) and then mixed for 10 min using a spatula. Then, 1 g of PTFE (Sigma Aldrich) was added, and the slurry has been mixed for another 10 min. At last, 2.75 g of carbon powder was added, and the overall content was mixed for an additional 10 min. The resultant mixture was then

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