



Re-use of carbon rods from used batteries as cathode for textile azo dye degradation in a microbial fuel cell

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ABSTRACT

Microbial fuel cell (MFC) has been used to treat various types of organic effluents using microbes as biocatalysts, producing electrical energy. The electrodes play a significant role in the MFC bioelectrochemical processes. The present study reports the use of carbon rods from used batteries as cathode material for textile dye degradation and decolorization. The concept of using carbon rods from used zinc carbon cell is novel and a green process of waste recycling in a MFC. Real textile dye effluent was loaded in the MFC anode. The MFC under investigation had produced a maximum power of 56.42 mW and with 90% reduction in COD over a time period of 600 h. The percent decolourization attained was 28.5%. The FESEM analysis of biofilm observed was opaque and thick sheets surrounded by a strong coat of possibly exopolysaccharide. Bio-film under optical microscopy, the microbes appeared to be isolated and bilobed. The results of this study indicate that the carbon rods from zinc carbon cell can be harvested as a MFC cathode for dye degradation and simultaneous power production.

Keywords: Microbial fuel cell; Textile dye; Used battery; Carbon rods; Wastewater; Degradation; Decolourization; Bioelectricity

1. Introduction

Urbanisation, population growth and environmental pollution have all grown tremendously with the rapid industrialization [1]. Textile and dyeing industries are found to be major polluters of the environment [2]. This industry consumes huge amounts of water in its manufacturing processes due to the wide spectrum of commercial dyes used in their processes [3–4]. Dyes possess table aromatic ring structures [4]. These aromatic structures make the dye molecules recalcitrant to bacterial degradation and it persists in the environment for an extensive period of time [5]. In addition, dye degradation products have been reported to be mutagenic, carcinogenic, toxic and teratogenic to both terrestrial and aquatic life [6]. Consequently, the decolor-

ization and degradation of dye wastewaters have become a great challenge [7].

Several treatment technologies are available for the degradation and decolorization of dye effluents [8–10]. However, there are constraints in the use of these conventional treatment methods and among them is high operational cost [11], and the need for secondary treatment of sludge [12]. Recently, advanced oxidation processes (AOPs) have received considerable interest in complete dye degradation [13]. However AOPs are not economically feasible due to its complex procedures involved [14]. Biological treatment methodologies are greener and low-cost. It is possible to completely degrade dye effluents using biological treatment methods [15]. For eg., anaerobic processes have proved to be a viable option for dye effluent treatment [16]. Microbial degradation and decolorization is a eco-friendly and an alternative to cost intensive chemical degradation

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procedures [17]. Microbial fuel cells (MFCs) have evolved as an option for microbial treatment to remediate dye effluents by degradation and decolorization with simultaneous energy production [18]. However the bio-electricity generation in an MFC is quite low for practical applications [19,20]. MFC was first applied in electro Fenton reactions for p-nitrophenol degradation and electricity generation with zero external energy supplied [21].

The ability of the MFCs to form a biofilm at the anode aided in the degradation of organic contaminants [22,23]. Electroactive microorganisms in MFC can serve as a biocatalyst to degrade the organics into electrons and protons. Electrons shuttle from anode to cathode via the circuit and combine with oxygen and protons to form water [24]. Therefore, electrodes have an important role in the efficiency of an MFC [25]. The most common electrode materials that are used traditionally as anode and cathode include carbon cloth, graphite and carbon paper [26].

Sophia and Saikant [27,28] studied the MFC for chromium removal using graphite sheets as electrodes. In recent studies, platinum electrodes were used as a cathode for MFC. However usage of platinum is not economical [29]. Studies have been highlighted on the assembly of the novel, cost-effective electrode materials and membranes to improve the MFC performance [30,31].

The present study uses carbon rod from a used zinc carbon cell as MFC cathode in achieving dye decolorization, degradation and simultaneous bio-electricity generation. The idea of using carbon rod from zinc carbon cell is aimed to improve the surface of the electrode, so as to increase the quantity of colonizing bacteria which in turn produces current. This study suggests, that the amorphous nature of the carbon rods from battery waste can generate maximum power in a microbial fuel cell. The electrode material can be used in any type of MFC due to its rigid size and shape, and cost-effective nature. The novelty of this study is that the carbon rod from used zinc carbon cell has been used first time in the MFC for real dye wastewater degradation.

2. Materials and methods

2.1. MFC setup and microbial media

Two chambered MFC was fabricated using Plexiglas with chambers working volume of 600 mL each separated by a cation exchange membrane, Nafion 117 (Sigma-Aldrich) [32]. Carbon brush was used as the anode and two carbon rods from used zinc carbon cell were used as cathodes. The total surface area of the cathode was found to be 16.5792 cm², which was similar to the area of cathode reported by Mashkour and Rahimnejad (2015) [33] using graphite rod, CNT/Pt, carbon cloth and carbon paper. The photograph and schematic representation of an experimented MFC are shown in Fig. 1. The whole MFC study was operated at ambient conditions with a neutral pH range of 6.5–7, maintained by phosphate buffer solution (PBS) in the anode compartment.

The experiment was conducted in batch mode at room temperature with an external load connected (10 Ω). Textile dye wastewater collected from a dyeing unit in Kancheepuram, Chennai was introduced into the anode enriched with pre-acclimatized activated microbial sewage sludge (pH 7.0)

containing substrate sodium acetate (1.5 g/L) in 50 mM PBS of (g/L) NaH₂PO₄·H₂O: 2.45 g, Na₂HPO₄: 4.58 g, NH₄Cl: 0.31 g, KCl: 0.13 g [34], 12.5 mL/L of mineral metal solution and 5 mL/L vitamin solution [35] and the cathode was amended with potassium ferricyanide in PBS solution [36].

The whole experiments with similar conditions were performed in triplicate with each study for a time period of 600 h. The parameters such as pH, conductivity, current and voltage were recorded during the run at definite time intervals. The anode was maintained in anaerobic conditions sealed with the rubber septa and cathode was aerobic for the entire study.

2.2. MFC experimental analysis

The current (mA) and voltage (mV) of the cell for each MFC run were recorded at specific time intervals using a fixed digital multimeter (Mastech M3900). The pH and conductivity of anolyte solution were monitored using a digital pH meter (Roy Instruments, India) and conductivity probe (Eutech Instruments, India). The percent dye decolorization was measured spectrophotometrically with Shimadzu, UV-2450.COD for the MFC anolyte was performed as per APHA the standard methods [37] to observe the dye degradation.

2.3. Optical imaging, FESEM-EDX analysis of biofilm

Biofilm optical microscopy (Zeiss-SteReo Discovery. V20) was investigated to visualize the microbes present on its surface. The collected biofilm from anode was fixed on a glass slide, washed with acetone followed by ethanol/methanol and then air dried. FESEM-EDX was performed as explained earlier [19] to observe the degree of microbial colonization for the pre-acclimatized biofilm anode after 600h of the reactor period.

3. Results and discussion

3.1. Power production

The present MFC study has shown results of dye color removal and voltage production. The voltage and current outputs of MFC experimented for the dye at an external resistance of 10 Ω was found to be 9.70 ± 1.92 mV and 0.97 ± 0.11 mA (Fig. 2). Theoretical analysis of cell emf may be several times higher than the experimental value obtained but it will not take into account the internal losses such as ohmic losses, activation losses, bacterial metabolic losses and concentration losses. In the present study, we are dealing with real complex dye wastewater which may not support theoretical calculations done under ideal conditions. The highest power density obtained during the run was 1146.8 mW/m². This was considerably higher than the MFCs that used platinum coated carbon electrode as cathode [38]. Study is in progress to find out how and why carbon rod from used zinc carbon cell act as better cathode, keeping all the other conditions the same. The maximum current density calculated in the present study was 11.2 mA/m². The higher current densities in this MFC study could probably be due to low external resistance (10 Ω) [39,40].

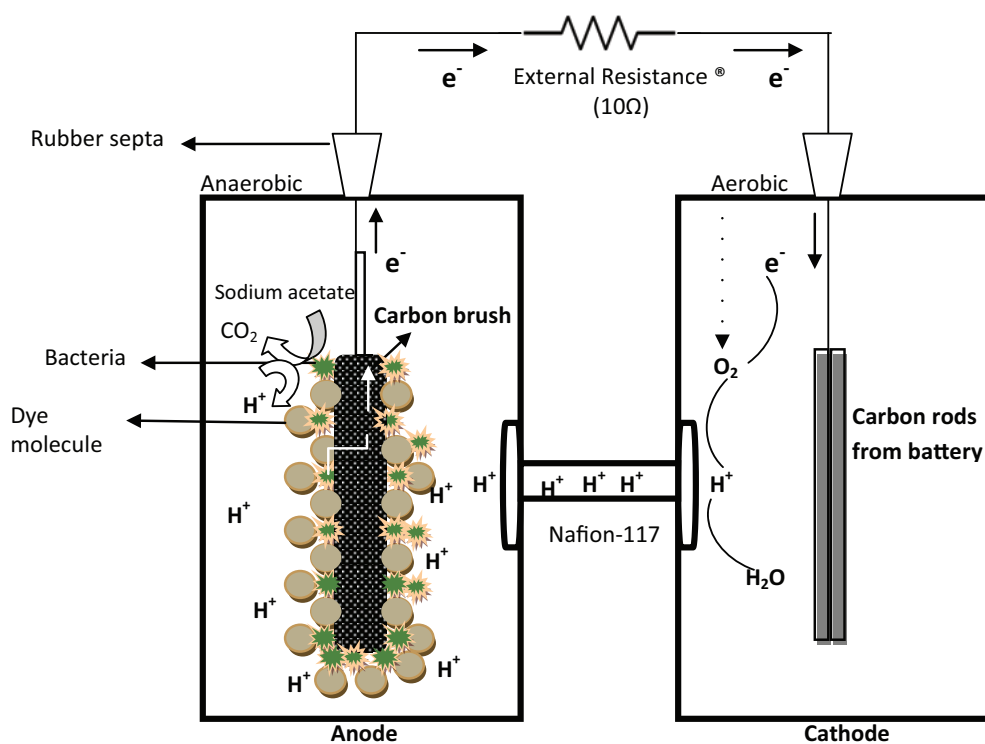


Fig. 1. Schematic and photograph of two-chambered microbial fuel cell with textile dye effluent as the anode substrate.

Highest power obtained during the MFC run in the present study using carbon rod from used zinc carbon cell, was 56.42 mW. This value is almost equal to the values reported by Wang et al. (2013) [41] with glucose as the substrate, carbon cloth cathode for electricity generation fuelled with the hydrolysis of enteromorphaprolifera by the MFC.

The present research indicates that the efficiency of MFC in electricity generation was improved by using carbon rod from used zinc carbon cell as a cathode. The obtained results also proved that mixed microbial consortium played an active role in the degradation of complex organics [42]. Research is in progress to find out the nature of the

bio-film and the anode respiring bacteria using biological and molecular techniques.

3.2. pH and conductivity

The value of pH plays a vital role in the physiognomies of textile wastes [1] and in generating the hydroxyl radicals during Fenton process [43]. The pH was monitored in the anode chamber of MFC with textile dye waste and is presented in Fig. 3. The pH of the anode solution varied from 6.0 ± 0.40 to 6.7 ± 0.60 . This variation in pH of the anode solution may be due to the protons produced in the anode

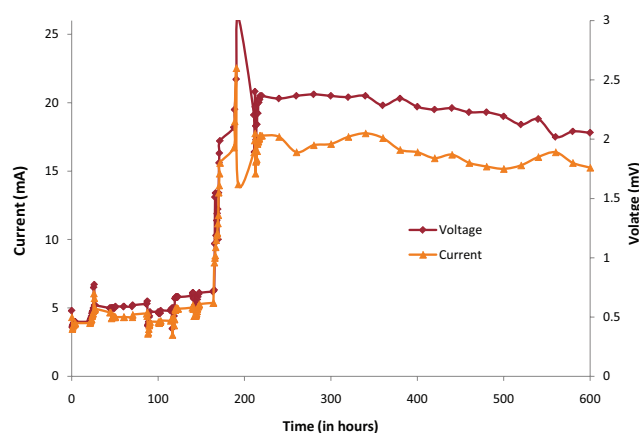


Fig. 2. Values of current and voltage obtained in a MFC with textile effluent dye.

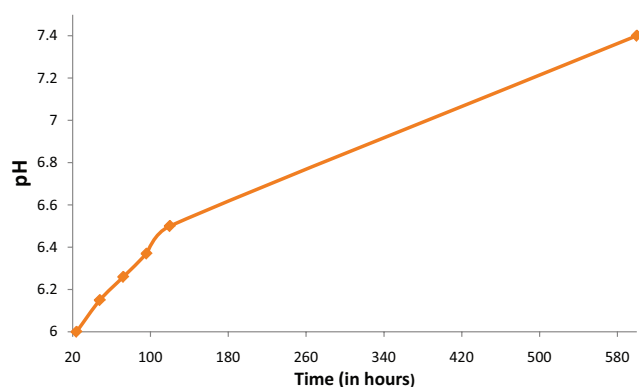


Fig. 3. Values of pH obtained in a dye MFC with textile effluent dye.

chamber and their movement from anode to cathode. The pH was observed to be close to neutral through the entire MFC run. This may be attributed to the anolyte buffering. The rate of degradation of dye effluent is found to be lower in the lower range of pH [44]. The value of pH has been observed to influence dye degradation and the optimum value for dye removal usually ranges from 5.0 to 7.0 [45]. This may also be supported by the fact that the optimum pH for bacterial growth is close to neutrality [46]. Bayoumi et al. (2014) [47] studied the bioremediation of textile wastewater using different bacteria in an MFC, where the optimum pH for decolorization was observed to be 7.0 and 90% decolorization. The pH value for dye decolorization obtained in our study was analogous to the pH reported by Prasad et al. (2013) [48].

Fig. 4 shows the conductivity of the anode solution decreased from 14.28 mS/cm to 6.80 mS/cm. The anode conductivity in the present study had decreased about 45%, while it was 41% for dye degradation using MFC studied by Khan et al. [49]. It is reported by Liu and Cheng [50] that the conductivity of real wastewater is typically low to uphold the internal resistance. As the conductivity of anode solution increased, the internal resistance decreased [51]. Most of the domestic wastewaters in MFCs

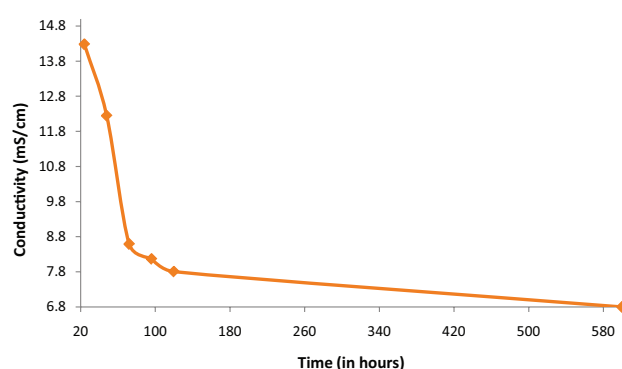


Fig. 4. Values of conductivity obtained in a MFC with textile effluent dye.

reported low conductivity with an approximate of 1 mS/cm which could increase the internal resistance of the system [52]. Therefore, low conductivity wastewater has become a great challenge to recover power production in BES systems.

3.3. Dye degradation and decolorization

The dye degradation in the MFC was monitored using COD reduction. Fig. 5 illustrates the COD removal in the MFC anode. The initial COD concentration of the raw textile dye was 1386.84 mg/L. High COD may be ascribed to high concentration of organics present in the effluent [53]. Similar results have been reported by Sindhi and Mehta, (2014) [54] using different industrial wastewater and their corresponding COD removal. The present study demonstrated percent COD removal of 90.6% which was higher than the COD removal (57%) of dye wastewater reported in literature [55]. This study shows 90% COD removal, which was higher than in reported in (85%) literature using MnO_2 catalyst as cathode [56].

The COD values of the treated effluent was found to be within the permissible limit of effluents [57]. With the decrease in COD, power densities increased for textile effluent dye. This may be due to the recyclable organic carbon of primary and secondary alcohols responsible for power generation [58]. Juang [59] reported similar performance of the MFC system with constant power output and COD removal efficiency.

The dye decolorization was monitored using the absorption spectrum of textile dye anolyte at room temperature using UV-VIS spectrophotometer in the range of 200–700 nm. The percent decolorization of dye in the MFC is presented in Fig. 6. The percent dye color removal calculated during the study was 28.5%. The increase in decolorization of the dye effluent may be due to the presence of active microbes responsible for dye decolorization and azo dye degradation. Khan et al. [55] reported that the spectra of Yellow Cibacron-2G azo dye band at 510 nm after the anaerobic treatment had decreased substantially in an MFC. There was no remarkable changes in color removal observed during the first two days of the study and gradually color removal increased after the 4th day.

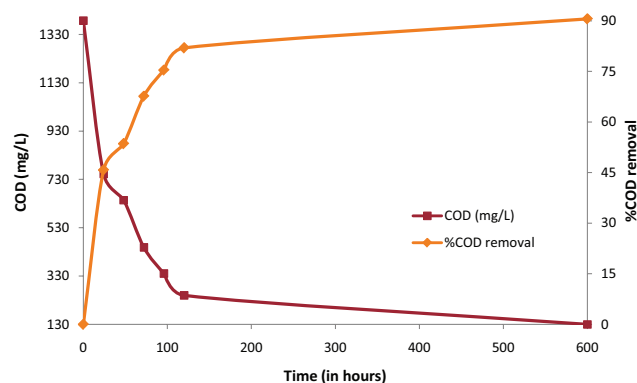


Fig. 5. Values of COD and percent COD removal observed in a MFC with textile effluent dye.

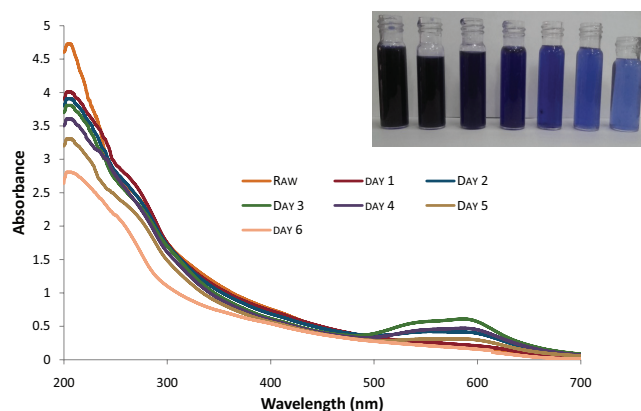


Fig. 6. Image of dye color removal and absorption spectra of the textile dye effluent in a microbial fuel cell.

The percent decolorization obtained using carbon rods from used zinc-carbon cell was compared to the literature reported (25%) by Khan et al. [60] in an MFC with an external resistance of 130 Ω . The decolorization efficiency of the present study may be due to the transfer of electrons within the anode compartment.

3.4. Biofilm optical microscopy

The anode bio-film image after 600 h of MFC operation was recorded in an optical microscope, FE-SEM, EDX are presented in Fig. 7. The optical examination of biofilm through a microscope demonstrated bacterial growth and proliferation on the anode. The image showed the microbes are isolated, bilobed with intricate ribbon structures.

FE-SEM image of the biofilm appears dense, sporadic colonization with coccus morphology. It also shows that the microbes that are opaque, made of thick sheets and may be bounded by a strong coat of exopolysaccharide [61]. Geetanjali et al. [62] have also reported the similar interpretation of biofilm. The possible presence of polysaccharide coating around the bacteria in the present study may be an advantage in the MFC. This may provide sites for positively charged species getting attracted and thus creating an ion-exchange medium [63]. Previous studies on biofilm had

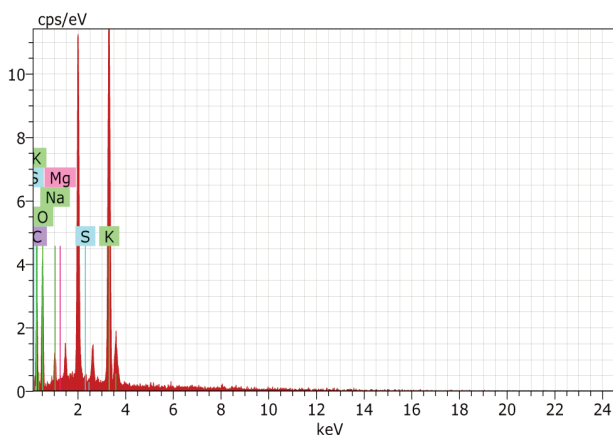
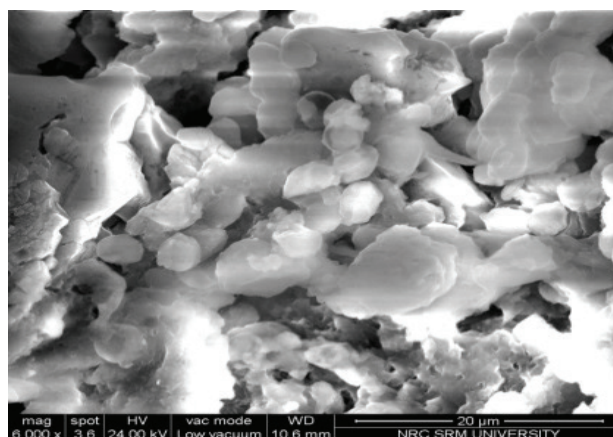
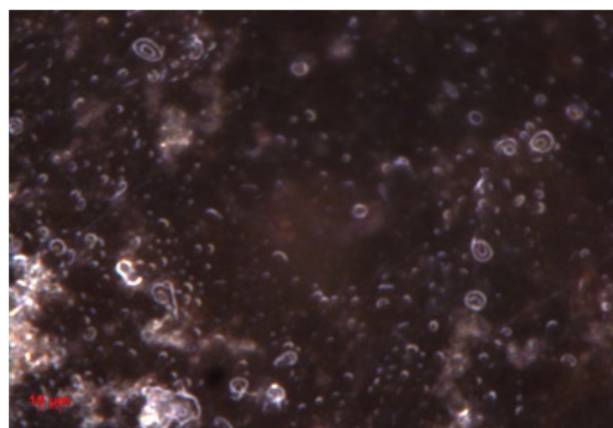


Fig. 7. Optical microscope, FESEM-EDX image of biofilm on the anode.

explained that the growth of bacteria in bioelectrochemical systems [48].

The biofilm was evaporated in a controlled environment to find out the salt deposits. The EDX of anode biofilm is shown in Fig. 7. The spectra revealed the foremost elements present in biofilm were Na, K, Mg, S, C, O. The EDX analysis has shown a high percentage of potassium. Among the above elements, sulfur, carbon, and oxygen were found to be intrinsic to the CEM polymers that are mainly induced by the bacterial growth [64].

4. Conclusions

Bioelectricity generation is possible during the decolorization and degradation of textile dye waste under the co-substrate in the double compartment MFC. The foremost study of using carbon rods from the battery waste for textile dye degradation as the cathode material had involved a green process for waste reuse. The advancement in electrical energy output was shown well under the presence of carbon electrode with the anode inoculated textile dye waste. Maximum power reported in this study was 56.42 mW in the presence of sodium acetate. The percent dye COD removal was 90% for a time period of 600 h. The percent dye color removal for the anode textile dye effluent was 28.5%. The carbon rod from used zinc-carbon cell as cathode experimented had produced a pH of 7.3. From the results shown, it can be seen that MFC technology is a viable alternative for simultaneous dye decolorization and degradation with energy recovery from realistic dye wastewater management. The predominance of cathode electrodes as the electron acceptor had favored high removal rate. The processed images of biofilm observed by FESEM and optical microscope in the present study had proved that the increased power densities were mainly accomplished by the bacterial growth. This provides the fundamental tool for identifying the physiological behaviors of bacteria and their interaction with MFCs. The future scenario of the intended approach towards MFC can be made useful as a low-cost wastewater treatment system.

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