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Development of Hybrid Microbial Fuel Cell for Waste Water Treatment System and Electrocoagulation

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ABSTRACT

The bio-electro chemical treatment systems (BES) are not only capable of producing electricity also treat the waste water; however the high cost poses hurdles towards their field application. In present work, microbial fuel cell (MFC) was constructed using low cost ceramic membrane for achieving the economy in construction and Zirconium dioxide (ZrO_2) was used as cathode catalyst in order to achieve high performance. MFC consists of eight chambershydraulically separated but electrically connected used for the secondary treatment of waste water while inner octagonal chamber was used to carry out the electrocoagulation. The results indicate that the Chemical oxygen demand (COD) removal was 75 ±4% during the first two weeks; however it was improved after fourth week with

90 ±5% COD removal. Maximum turbidity removal for the controlled setup was observed to be 71% while for the uncontrolled setup it was only 48%. Hence, the proposed device has potential benefit of achieving secondary and tertiary wastewater treatment simultaneously along with energy recovery.

Keywords

Chemical Oxygen Demand; Ceramic Membrane; Electro-coagulation;Microbial Fuel Cell; Zirconium dioxide

1. INTRODUCTION

Many research and technological advancements have been made in the area of renewable energy sources and technology. This is due to the rapid exhaustion of the fossil fuel based energy sources which continuously increase in cost i.e. oil. Another important issue face by humankind is the environmental pollution by wastewater generated from domestic and industrial activities. The wastewater should be treated before discharging into the environment. In this regard, microbial fuel cells (MFCs), that are one kind of denominated bio-electrochemical reactors, are promising and efficient technology for the wastewater treatment that converts the stored chemical energy in the chemical bonds of organic matter to the electrical energy, as well as many applications that employ the bio-electrochemical reactions and microorganisms as biocatalysts. Although, considerable progress has been achieved in the performance of a MFC in the past ten years, one of the main challenges for commercializing scalable MFCs is the high cost and low mechanical strength of the separator materials used for fabrication of this device.The microbial fuel cell is a bio-electrical system in which bacteria is used to convert organic material into electricity. The fuel cell itself is made up of electrogenic parts; the anode, the cathode, the proton exchange membrane and the external circuit. The electrons are pulled out as released energy during the oxidation process and into the electron acceptor via an external circuit. The protons pass through the ion/ proton exchange membrane and react with the electrons during the reduction process in the cathode thus completing the circuit. This simple process which is common and found in most fuel cells i.e. battery cells, hydrogen fuel cells can be optimized for an efficient current generation. The exploration of various materials used in electrodes that balances efficiency and cost-effectiveness is the key to the potential large scale use of MFC particularly in wastewater treatment plants.

Several designs were used by the researchers to obtain high power density such as single chamber MFC, up flow MFC with an interior cathode, flat plate microbial fuel cell. Different substrates are used for electricity generation viz, synthetic Waste water, industrial Waste water, domestic Waste water, etc. The performance of the reactor can be improved by decreasing the internal resistance of the system, using efficient electrode material and fabricating efficient system architecture, which can provide improved environment to bacterial community to work efficiently (Sonawane et al., 2013). Ceramic membrane turn out to be a material that can be used in microbial fuel cell because of their low cost production and better strength, thus can be used an alternative for costly material. Many researchers have practiced the use of ceramic in microbial fuel cell in order to improve the performance and reduce fabrication cost. Ceramics are still used to this day for the electrochemical treatment of wastewater, particularly by transforming pollutants into non-toxic materials (Winfield et al., 2016).The ceramics can provide stability, improve power and treatment efficiencies, create a better environment for the electro-active bacteria and contribute towards resource recovery. MFC found to be promising system for organic removal and electricity generation. However, its application for electrocoagulation using multi-electrode system is not yet reported so far. Hence, this paper aims at designing and evaluating multi-electrode bio-electrochemical system for secondary and tertiary treatment, simultaneously. Also it presents the development of cheaper zirconium catalyst for oxygen reduction.

2. MATERIALS AND METHODS

2.1. Fabrication of Set up:

Multi-electrode Microbial Fuel Cell is fabricated using transparent acrylic sheet of thickness 5mm in the form of Octagon with a height of 20 cm (Fig. 1). The reactor consists of eight square chambers of size 10.5cm x 10.5cm with height of 20cm and volume of 2.205L each. The inner octagon chamber with each side measuring 11cm and height of 20cm with a volume of 11.68L will be used to carry out the electro coagulation (Fig. 1). On each face of the outer rectangular chamber, three rectangular windows of dimensions 7.5 cm x 10cm are created for placing ceramic membrane in turn will support for membrane electrode assembly at cathode. The windows on the reactor can accommodate a total cathode surface area of 600cm^2.These windows are covered with low cost effective ceramic membrane that acts as a separator (Fig. 2). The outer surface of the cathode will be covered with the conductive carbon ink prepared using carbon powder (Vulcan-X72) along with the catalyst called Zirconium dioxide to accelerate the reaction. The Square chambers provided are mainly used for the secondary wastewater treatment while the octagonal is used for electro coagulation.

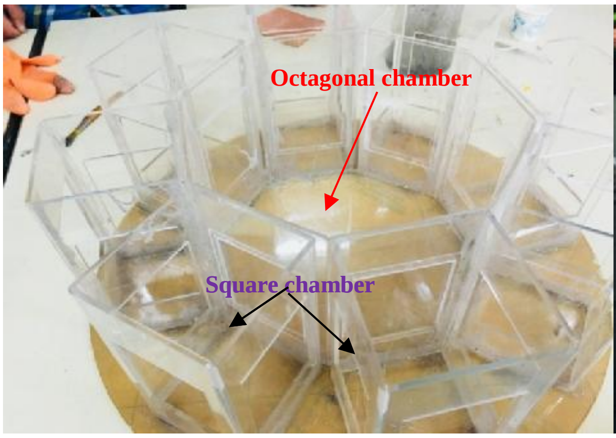

Fig 1: Set up

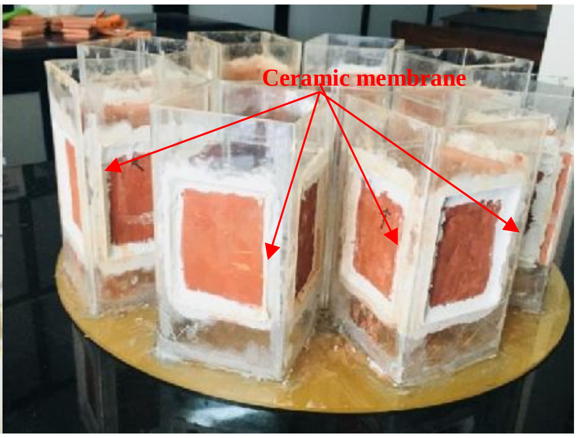

Fig 2: Fabrication of MFC using Ceramic membrane

2.2. Preparation of Anode:

The materials used as anode electrodes must have several specific characteristics for improving interactions between the EA biofilms and the material surface. The most important characteristics are: i) electrical conductivity; ii) resistance to corrosion; iii) high mechanical strength; iv) developed surface area; v) biocompatibility; vi) environment friendly and vii) low cost.

Carbon felt of thickness 0.5cm was used to fabricate the anode electrode of dimension 7.5cm×10 cm as shown in the (Fig. 3).The objective of using carbon felt is to provide large surface area that enhances the microbial adhesion, which results in well-established bio-films.Carbon felt held in position with the help of steel mesh so it should stay in contact with ceramic membrane. Carbon felt sheet along with the steel mesh is inserted in all the eight chambers. Each of the carbon felt anode electrode is further connected to the external load with help of tungsten wire as shown in (Fig. 3).

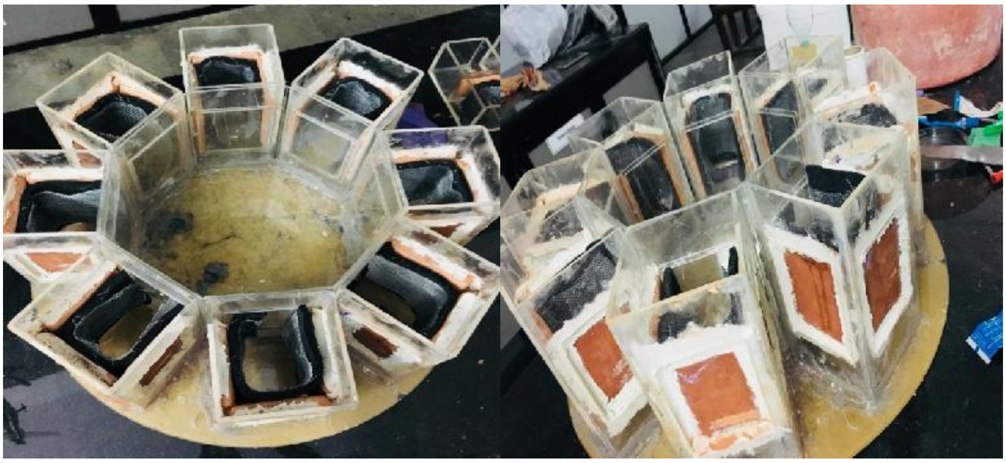

Fig 3: Anode Configuration

2.3. Preparation of Cathode:

The oxygen reduction reaction that is taking place at the cathode is often the limiting reaction of the MFCs. The oxygen reduction reaction (ORR) in neutral media can be facilitated by the utilization of enzymes, microbes or abiotic catalysts. Fine carbon powder ink made up of Carbon powder (Vulcan X-72), Catalyst ZrO_2, Polyvinyl Alcohol Solution and Acetone. Interestingly, in the field of MFCs, the performances of catalysts incorporated into the cathode are better studied than the kinetics studies of the catalysts itself. Catalyst option based on carbonaceous materials seems to be viable for low cost practical applications, lowering the capital costs and having relatively high and stable performance. The characteristic of cathode catalyst were analyzed using X-ray diffraction analysis (XRD) at TIFR and Scanning electron micrograph (SEM) at IITB

2.3.1 Cathode catalyst (Zirconium dioxide)

Catalyst used in preparation of carbon ink and in order to increase the organic load rating is Zirconium dioxide (ZrO_2). The material required for production of ZrO_2 are Benzilic Acid – 18.44g, $NaHCO_3$ – 6.72g, $ZrOCl_2 * 8H_2O$ – 12.88g (Fig. 4). While the procedure followed for the production of Zirconium Dioxide-

1. In500ml beaker takes Benzilic acid and adds 100ml of diluted water to this 6.72g of $NaHCo_3$ is gradually added.
2. In 250ml beaker take ZrOCl2 and add 50ml of diluted water. This solution is gradually added to first solution and yields a white precipitate.
3. The white precipitate thus obtained is washed three times in 600ml of diluted water by stirring for one hour to remove the reagents.
4. Once the washing is done properly the precipitate is then dried in vacuum for 12hrs and dried in oven below 100°C.
5. Pyrolysis is carried out at 588°C in Muffle Furnace.

Fig 4: Preparation of cathode Catalyst (ZrO_2)

2.4 Turbidity Measurement:

The amount of suspended solids in water bodies is an important factor while considering the water quality. These solids that often include silt, clay, algae, organic matter, and other minute particles, obstruct the transmittance of light through the water and impart a qualitative characteristic known as turbidity. Turbidity is often related to the surface water conditions and changes in turbidity are therefore indicator of changes in environmental condition. Turbidity is a measure of the cloudiness of water. The higher the turbidity, the harder it is to see through the water. Turbidity measurements are reported in nephelometric turbidity units (NTU).Turbidity can be considered as a measure of the relative clarity of water. Although it's not the direct measure of the suspended particles in water but it is measure of the scattering affects these particles have on light. The higher the intensity of the scattered light, the higher will be the turbidity. The water Quality is influenced by many parameters however turbidity is given noteworthy importance because it's simple and undeniable indicator of water quality. High turbidity can indicate higher concentration of bacteria, nutrients, pesticides, or metals. Any sudden change in turbidity may indicate the presence of a new pollution source in natural water.

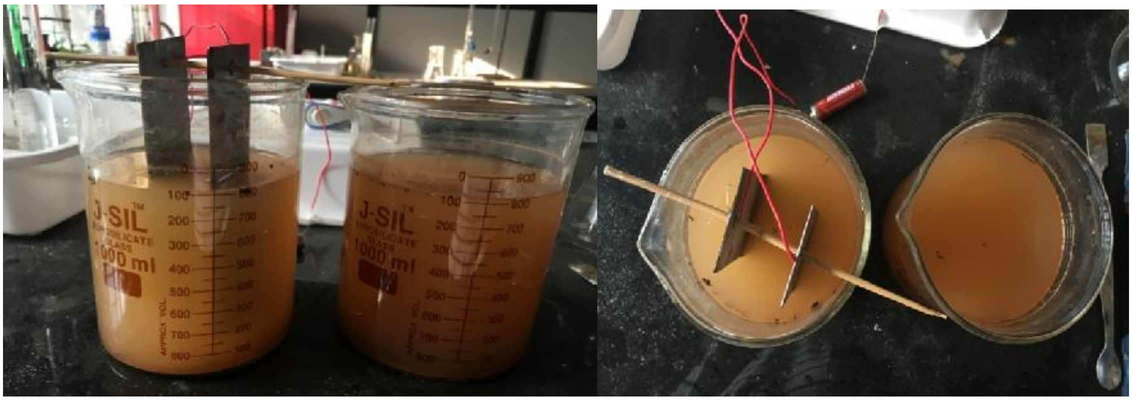

Fig 5: experimental set-up for Turbidity Measurement

A Synthetically Prepared turbid water of 1300 NTU was used to measure the efficiency of the prepared system. There were two samples made of same turbidity one of the sample was allowed to settle the suspended solids naturally while the other sample was treated by connecting a battery to iron plates. To stimulate the performance of MFC battery of 2.5V(Fig. 5) was used to develop potential between the two iron plates and the water samples were collected at regular interval of 1Hr from controlled as well as uncontrolled set up. The Turbidity was measure using turbidity meter(Fig. 6).

Fig 6: Turbidity meter

3. RESULTS AND DISCUSSION

3.1 Characterization ofZirconium catalyst

Zirconium oxide is characterized to be the only one metal oxide which possesses explicitly four chemical properties on the surface: acidic and basic properties and oxidizing and reducing properties**.**The high ion exchange capacity and redox activities make it useful in many catalytic processes as a catalyst. ZrO^2has three well-defined crystal structures/phases, that is, cubic (c- ZrO^2), tetragonal (t-ZrO^2), and monoclinic (m-ZrO^2), under normal atmosphere and at different temperatures.The prepared annealed samples were characterized for their formation, structure, morphology, and elemental composition using X-ray diffraction analysis (XRD), scanning electron micrograph (SEM).Figure 7 shows the XRD pattern of the crystalline zirconia powder heat treated at 700°C for 1 hour.

3.2 X-ray Diffraction

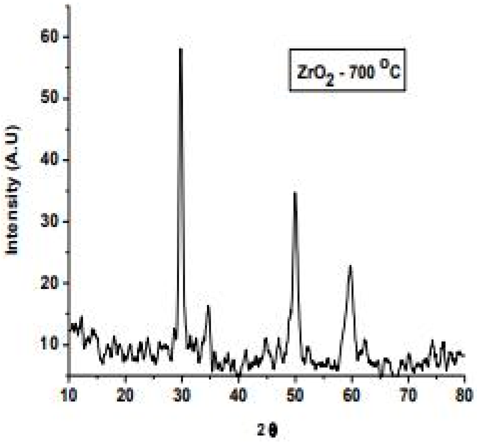

Fig 7: XRD pattern for Zirconium oxide

There are four strong diffraction peaks at $2\theta = 29.87^{o}$, 34.40^{o}, 49.97^{o}, and 59.69^{o}, which can be attributed to the formation of the zirconia [9-14]. It should be noticed that, when Zirconia powder is heat treated at 4000 and 5000 for one hour, XRD studies donot shown any prominent crystalline peaks. As the heat treatment temperature increases to 7000, the crystallinity is formed in the synthesized zirconia powder. This is being more important in the case of the organic precursors used for the preparation of inorganic zirconia. The average size of the zirconia particles can be calculated from the full width at half maximum (FWHM) values of the diffraction peaks using Debye-Scherrer formula.

$$D = \frac{0.97\lambda}{\beta \cos\theta}$$

Where, 'D' is the average crystallite size, 'λ' is the wavelength of the x-rays used (CuKα of radiation $\lambda = 0.154$nm), 'ϴ' is the diffraction angle and 'β' is the FWHM. The size of the nanoparticles are estimated at $2\theta = 29.82°$, 49.97^{o}, 59.69^{o}. The average crystalline size of the synthesized zirconia is 253.93A° (25.39nm).

3.3 Scanning electron microscopy analysis (SEM Test)

SEM analysis of ZrO_2 show well defined rod like structures in the size range 42.8nm to47.8nm which is well within the desired size range of 1-100nm (Fig. 8 and Fig. 9).The images for the size of zirconium dioxide is attached below and this is as per the temperature at which the benzilate was heated to obtain ZrO_2 at 590 °C. SEM images were recorded for studying the microstructure and morphology of the catalysts. These SEM images showed that the synthesized catalysts were in nano-size. Highly inter-connected pores of sizes lesser than 1lm were observed in mostof the conditions. Well-separated particles with irregular shapewere observed for Zr-based compounds. Porous structures with interconnected network poreswere also observed in zirconia. EDX analyses also confirmed the presence of Zirconia incomposite catalysts. The peaks observed during EDX analyses showed high percentage of Zr with no impurities in the synthesized catalysts (Fig. 10).

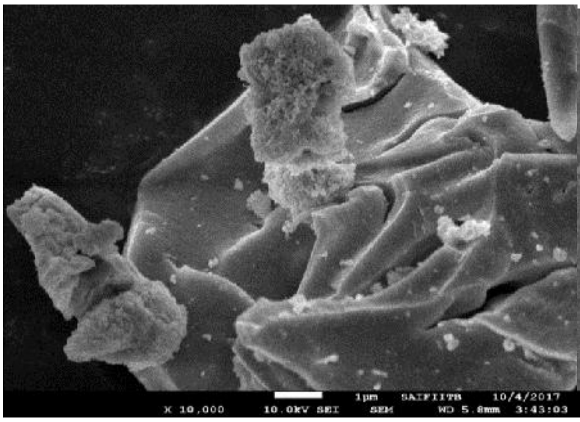

Fig 8: SEM of Crystal form ZrO_2

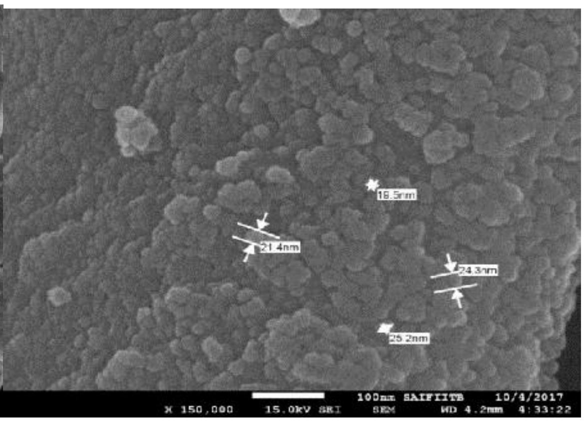

Fig 9: Nano-size of ZrO_2

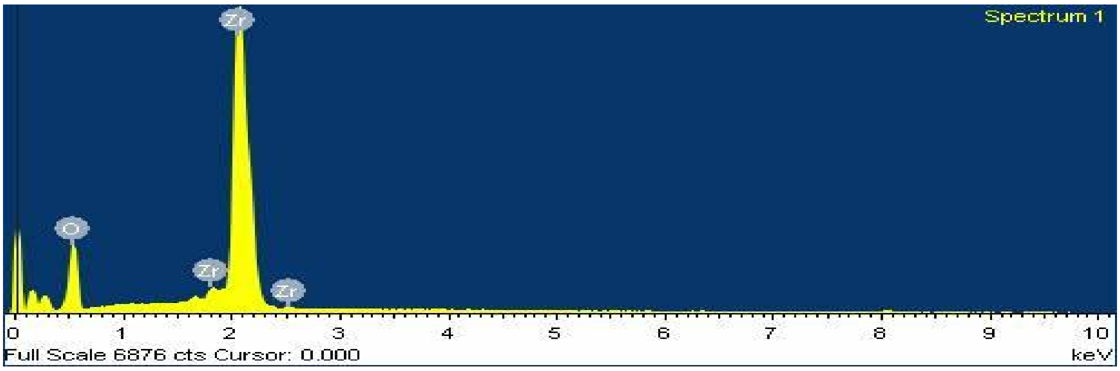

Fig 10: EDX image

3.4 Organic Removal

To check the efficiency of bio-electro chemical reactor for secondary waste water treatment synthetic waste water of 500 mg/l COD was prepared usingsodium acetate as carbon saluteinitial and final COD were measured(Ghangrekar and Jadhav, 2009). In cycle 1 the COD removal was 70% While in Cycle 2 and Cycle 3 the COD removal was maintained up to 90 ±5% (Fig 11). The results of the study indicate that the removal was 75± 4% during the first two week. However, there is a drastic change in COD removal of 90 ±5% observed after 4th week. This was due to better growth of biofilm on anode results in increase in redox process. Thus it can be combining that the present MFC system can be applied for treatment of waste water without any difficulty.

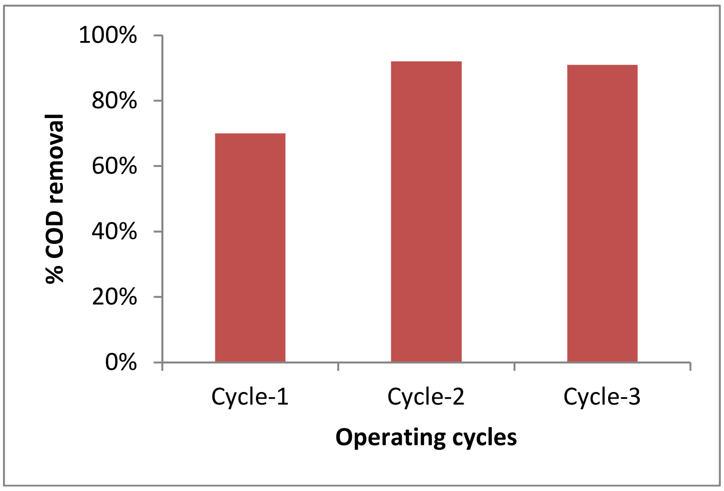

Fig 11: COD removal cycle

3.5 Distinguish between Gravity settling and electro-coagulation using MFC

As stated above there were two samples taken out of which one sample was treated using gravity to remove the suspended solids from the water while the other sample was treated by generating potential difference between two iron plates. One of the samples was controlled while the other one was not controlled.

1. Gravity Settling (Uncontrolled Setup)

Settling is the process by which particulates settle to the bottom of a liquid and form a sediment. Particles that experience a force, either due to gravity or due to centrifugal motion will tend to move in a uniform manner in the direction exerted by that force. For gravity settling, this means that the particles will tend to fall to the bottom of the vessel, forming a slurry at the vessel base. For settling particles that are considered individually, i.e. dilute particle solutions, there are two main forces enacting upon any particle. The primary force is an applied force, such as gravity, and a drag force that is due to the motion of the particle through the fluid. The applied force is usually not affected by the particle's velocity, whereas the drag force is a function of the particle velocity. For a particle at rest no drag force will be exhibited, which causes the particle to accelerate due to the applied force.

2. Electrocoagulation (Controlled Setup)

The development of electrochemical processes for wastewater treatment, remediation and disinfection has become a viable option in the past two decades.Many types of industrial wastewater have been found to be highly saline and can be treated by electro-chemical methods. Wastewater, which is polluted with heavy metal compounds, is usually treated with the use of adsorption, flotation and physio-chemical techniques such as electrocoagulation (EC). In general, electrocoagulation (also called electro flocculation) is a process that is used for water and wastewater treatment that destabilises finely dispersed particles.The EC technique uses a direct current source between metal electrodes immersed in polluted water, where sacrificial anode electrodes (aluminium or iron based) generate coagulants that destabilise contaminants. In addition, the cathodeproduces

OH ions generated in the reduction reaction. EC is an alternative method to conventional (chemical) flocculation because it avoids using chemicals, such as chloride or sulphate.

3.6 Performance Comparison of Controlled and UncontrolledCoagulation

In the present study, it was observed that after period of two and half hour the turbidity removal for electrocoagulation was 47% while for normal gravity settling it was 24% (Table 1). However, the rate of removal for turbidity due to normal settling was decreased till 43% after that there wasno marginal decrease in the rate of turbidity upto 47% this indicate the particle which are of colloidal nature remain in diluted state and particle of larger size are already settled. While the rate of removal for controlled setup was 48% this removal was not attractive since the number of Fe+3 ion were not dissolved into the solution, After three hours the rate of removal was 60% and the turbidity removal was continued till 71% there was a constant reduction in turbidity that was observed (Fig. 12).

Table1:Turbidity Removal in NTU

Time (Min)	Controlled Setup	Percentage	Uncontrolled Setup	Percentage
0	650		650	
30	363	44	534	17
60	338	48	492	24
90	290	55	421	35
120	278	57	390	40
150	267	59	388	40.3
180	257	60	367	43
210	241	62	361	44
240	221	66	355	45
270	200	69	350	46
300	190	70	347	46.6
330	185	71	340	48

From the result it was observed that, at initial stage the rate of turbidity was 650 NTU; however with increase in time more number of Fe^{+3} iron are passed through the controlled solution with iron plate connected to battery where the particles settle down and turbidity is reduced. While in the normal settling sample the turbidity did reduce to a certain extend however after a point of time the decrease in turbidity was stable.

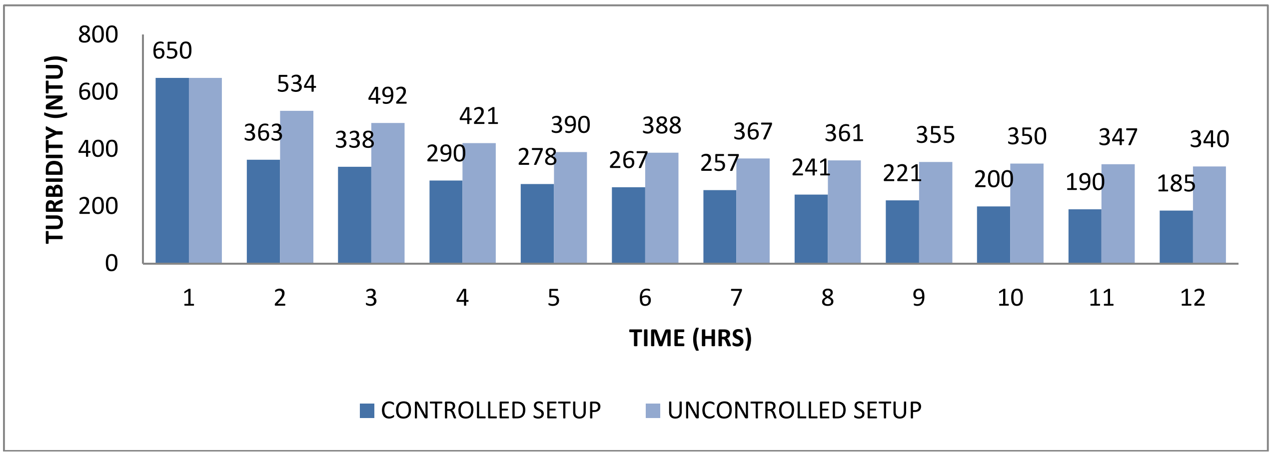

Fig 12: Bar Chart showing turbidity removal

4. CONCLUSION

Development of hybrid reactor in the bio electro chemical system (BES) is an attractive method to provide uniquesolution to waste water treatment and energy recovery.In this study, performance of an integrated hybrid MFC was evaluated for simultaneous secondary and tertiary treatments.Multi-electrode MFC systems with eight anode-cathode pairs with ZrO_2ascost effective cathode catalyst were used to enhance the rate of reduction reaction. The results of study demonstrate effective organic matter removal from waste wateralong with turbidity removal using electro-coagulation. This is the first study that claims the feasibility of hybrid multi-electrode MFC for simultaneous secondary and tertiary treatments. Usually electrocoagulation is carried out by applying current externallyfrom the outer source while in this research the current is generated within the system.Such low cost MFC could provide sustainable solution for wastewater treatment with electricity production and can take this technology a step ahead towards commercialization.

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