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Authors' contributions

This work was carried out in collaboration between both authors. Both authors read and approved the final manuscript.

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Original Research Article

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ABSTRACT

The interest for improving the quality of coffee production has led to the increased use of wet processing so as to highlight the quality and aroma of the beverage. The coffee pulping process has been causing environmental problems at the local level not only due to the consumption of water, but more due to the discharge of effluents with large volumes of organic waste. The wet processing generates enormous quantity of wastewater which causes irrepairable damage to the receiving water bodies. Indispensable. However, physico chemical treatment method coupled with biological treatment as proved effective treatment mode for safe and proper disposal of the wastewater generated. In this sense, the objective of the present study is to evaluate the efficacy of electrocoagulation batch reactor as pretreatment for anaerobic sequencing batch reactor to treat coffee processing wastewater.

Keywords: Coffee processing; electro coagulation; anaerobic; sequencing batch reactor.

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1. INTRODUCTION

One of the driving forces of the economy in India is agriculture. In India, coffee is one of the main agricultural products and is traded on the international market. Coffee is a plant a member of the genus Coffea in the family Rubiaceae. Caffeine is one of a class of xanthine compounds found in a long list of plant products, including tea leaves, cocoa beans and coffee beans. The world drinks about 2.25 billion cups per day - the United States alone drinks 1/5th of this. The first coffee houses opened in Constantinople in 1554. Coffee was introduced into North America in 1668. A pilgrim from India named Baba Budan brought the first germinal seeds from Mecca to Mysore around 1600. Currently India rank sixth in the world in coffee production, India grows about 3 lakh tonnes annually. In India, Karnataka is the largest coffee growing and coffee pulping state in India accounting for about 72% of total coffee produced. More than 3000 coffee estates located in the districts of Chikkamagalur, Kodagu, Hassan and Mysore carry out pulping (wet processing that needs huge quantities of water as well as discharge equally large quantities of water). It is estimated that nearly half the coffee grown here (40-50%) is processed by wet pulping method to yield a 'superior' quality product compared to the dry processing option also available.

Coffee pulping is subjected to mechanical and biological operation in order to separate the bean or seed from the exocarp, mesocarp, and endocarp. Much of this pulp is dumped in soils or water courses where its polyphenolic and recalcitrant substances cause serious damage to the terrestrial and aquatic ecosystems.

One of the wastewater treatment systems that have recently demonstrated good potential in biological process for organic matter and nutrient removal is the Sequencing Batch Reactor (SBR). The SBR is a batch reactor where in anaerobic, aerobic and clarification phases are achieved in a time sequence of continuous flow system. The SBR, in addition to organic carbon and phosphorous removal is also capable of nitrification and denitrification.

Recently, attention has turned to the electrocoagulation treatment of wastewaters, in which recalcitrant compounds are destroyed by direct or indirect anodic oxidation based on the generation of hydroxyl radicals, ozone, etc. They

claim complete degradation of phenols and total decolorization treatment and maximum reduction of COD.

1.1 Justification of Problem

Most of the industries like distillery, sugar, paper, pesticides, pharmaceutical, electroplating units, olive mill wastewater etc., have high strength organic waste in their effluents, Also there are certain chemical industries like dyes, pigment manufacturing, polymer textiles, pesticides etc., wastewater contains organic whose compounds (phenol, sulphonate derivatives viz... benzene substituted and naphthalenesulphonates, aromatic sulphonates), which are either slowly biodegradable or resistant to biological treatment. These refractory pollutants cannot be readily removed, usually are recalcitrant and not amenable for conventional biological treatment process and hence may depress the process efficiency. Sometimes it is difficult to achieve the removal of COD and BOD parameters within the stipulated standards as the initial COD and BOD concentration are very high as in the case of distillery and tannery wastewater. Distillery effluent has a 70,000 mg/L to 80.000 mg/L COD and moreover biological treatment process usually do not adequately remove color from textile industry wastewater.

Hence, to enhance the efficiency of the biological processes for industrial wastewater treatment, an effective pre-treatment process is always in demand to destroy these recalcitrant substances.

To overcome the above problem it is necessary to investigate alternate wastewater treatment method. The application of electrocoagulation techniques started with production of high volume chemicals, extraction and refining of metals. Today electrocoagulation technique such Electrodyflourdation, as Electro-Oxidation, (Flocculatation), agulation Electroco Electrocoagulation Electrodialysis. Direct process, Indirect Electrocoagulation process, Electrocoagulation Activation process etc., are emerging out as practical technologies replacing conventional regents in physicochemical methods of wastewater treatment. Electrocoagulation approach can be useful to degrade recalcitrant industrial wastewater or markedly enhance the biodegradability and biocompatibility in the treated effluents.

The electrocoagulation technologies offer simple, efficient and cost effective solution of

environmental problems and in certain application superiority over conventional clean up process. The superiority is due to the fact that by itself electrocoagulation methods do not require any external reagents for pollution treatment.

Electrocoagulation technology has shown to be a promising method for the destruction of organic pollutants in wastewater; there is no need for adding extra reagents. In addition, the high selectivity of the electrocoagulation process prevents the production of unwanted-by-product. The reaction can be carried out at room temperature and at normal pressure and be easily terminated by cutting off power supply.

2. MATERIALS AND METHODS

2.1 Wastewater Source

Coffee pulping wastewater was collected from pulping units of Coffee estates located in Chikkamagalur and Mercara districts. Coffee grown in this region is processed using the wet method, wet processing diminishes the bitterness and increases the acidity. This process gives rise to one of the smoothest, high-quality coffee. Coffee pulping wastewater was collected in polyethylene container and preserved in refrigerator at 4° C.

2.2 Characteristic of Raw Coffee Pulping Wastewater

The initial characteristics of coffee pulping wastewater collected from selected source is as shown in Table.1 The sample was collected from Chikkamagalur and Mercara estates. The wastewater is characterized bv hiah concentration of COD, solids, nitrogen and phosphorus with low pH. Since the wastewater contained high concentration of COD and nutrients the treatability studies of coffee pulping wastewater were carried out in laboratory scale Anaerobic Sequencing Batch Reactor (ASBR). The ASBR was operated with the five operating phases which include Fill, Anaerobic, Aeration, Settle and Decant phases.

Characteristic of wastewater collected from the processing units of selected source is as shown in the Table 1.

2.3 Bioreactor Setup

The experimental system used was composed of two individual reactors operated in sequencing batch mode under anaerobicaerobic microenvironment conditions with 24-hour cycle. In SBR each cycle consisted of a 17-hour anaerobic phase; an aerobic (react) phase 6 hour and 1 hour for settling, decant and fill phases. The SBR system was inculcated with the seed culture of cow dung with 300 ml in 1000 ml tap water. Cow dung was used as seed material since, cow dung is rich in heterotrophic bacteria which is responsible for denitrification process. The SBR was initially feed with raw coffee pulping wastewater in the phase-1 and later in the phase-2 coffee pulping wastewater was diluted to 1:4 and converted to Anaerobic Sequencing Batch Reactor (ASBR). ASBR was operated with electrocoagulation pretreated coffee pulping wastewater, as a feed solution.

Table 1. Characteristics of Coffee Processing Wastewater

Parameter	Concentration
рН	4.56 – 5.04
COD, mg/L	3872 – 4980
BOD, mg/L	152
Total Solids, mg/L	4758
Total Dissolved Solids, mg/L	4208
Volatile Solids, mg/L	1188
Ammonia -Nitrogen, mg/L	60 - 64
Nitrate -Nitrogen, mg/L	21 – 24
Phosphorous, mg/L	22 -24
Chlorides, mg/L	63.9 – 72
Sulphates, mg/L	4.42 – 4.5
Conductivity, µmho/cm	382.6 – 390.6
Alkalinity, mg/L	90 – 100
Total Hardness, mg/L	801 – 808
Calcium Hardness, mg/L	60 – 63
Magnesium Hardness, mg/L	720 – 748
Aluminum, mg/L	1.26 – 1.37
Cadmium, mg/L	0.18 – 0.21
Chromium, mg/L	0.20 – 0.26
Copper, mg/L	0.17 – 0.18
Iron, mg/L	6.3 – 6.83
Manganese, mg/L	0.63 – 0.67
Nickel, mg/L	0.42 – 0.45
Lead, mg/L	0.22 – 0.27
Zinc, mg/L	0.53 – 0.54

2.4 Anaerobic SBR

An anaerobic reactor of working volume 2 L with cow dung as a seed culture was used to treat the electrocoagulation treated coffee pulping wastewater. After electrocoagulation treatment the coffee pulping wastewater was fed to the reactor and kept under anaerobic condition for 23 hours. Further, 50 % of the reactor working volume was decanted everyday followed by addition of the same volume as feed. In anaerobic phase denitrification takes place, nitrate nitrogen will get converted to nitrogen gas, as a result nitrate concentration will be low. COD is utilized by heterotrophs as source of energy and thus COD decreases. On alternate days samples were collected to analyze the parameters like COD, ammonia nitrogen, nitrite nitrate and phosphorus. The operation of ASBR is as shown in the Plate 1.

2.5 Electrocoagulation Reactor

Electrolysis was conducted using aluminum electrodes in a glass beaker of 1000 ml capacity. The reactor was operated under completely mixed conditions, facilitated by a magnetic stirrer provided at the bottom of the reactor to avoid concentration gradients. The anode and cathode were positioned vertically and parallel to each other. The operation set up for EC is as shown in plate 2 and the bio reactor set up for maintaining anaerobic condition is shown in the plate 1.

2.6 Electrodes

Typically, EC equipment consists of DC-powered cathode and anode electrodes, which are partially submerged into a tank that contains a contaminated solution (Plate 2). Most often used shape for electrode material is rectangular shaped, while the shape, size and number vary [1]. The most common metals used to manufacture EC electrodes are Al and Fe because they are cost effective, widely available and non-hazardous [2,1,3]. The EC system can be operated in batch or continuous modes by either treating a fixed volume of waste effluent per process cycle or treatment of a continuous flow of the waste stream, respectively.



Plate 1. Operation of Anaerobic Sequencing Batch Reactor during Anaerobic Phase



Plate 2. Operation of Electrocoagulation reactor

Electrode assembly is the heart of the treatment facility and hence, appropriate selection of its material is very important. Hence, in the present study aluminum is selected as anode material. Aluminum is selected as it is cheap, readily available and proven effective. The description of electrode used is as follows:

- Aluminum plates of dimension 5 cm x 5 cm were used as anode and cathode.
- Weight of the two electrodes is 4.5513 gm and 4.6497 gm respectively.

2.7 Power Supply

The instrument Testronix 35 D, Dual DC regulated power supply is used to maintain current and voltage across the electrode system by means of copper wires.

2.8 Experimental Methodology

electrocoagulation experiments The using aluminum electrodes were conducted in a glass beaker of constant working volume of 1 L. The EC reactor set up is as shown in Fig 1. The anode and cathode were placed at a fixed distance of 1 cm to 2 cm apart. A 16 V power supply was maintained using DC power supply. Initially the raw coffee pulping wastewater was diluted (250 ml in 1000 ml) and added to the reactor. The electrocoagulation treatment was given for period of 2 hour. The foam which was formed on the top of the reactor was removed and the treated sample was filtered through filter paper, then the sample was analyzed for parameters like COD, nitrate nitrogen. ammonium nitrogen and phosphate.

2.9 Process Monitoring and Experimental Methodology

During the study, the parameters of concern was COD, ammonia nitrogen, nitrate nitrogen, phosphate and solids were measured routinely. The samples were collected at the end of anaerobic and aerobic phase along with feed in each cycle. The samples were filtered through ordinary filter paper before analysis. The procedures followed to analyze the parameters of concern were according to the standard methods (1992, 1998) [4-6]. The methodology adopted to analyze various parameters of concern are tabulated in Table 2.

3. RESULTS

3.1 Performance of Anaerobic Sequencing Batch Reactor (ASBR)

Since COD. nitrogen and phosphorus concentration in the wastewater was high an attempt was made in this study to treat the wastewater in a ASBR of working volume 3 L. In the ASBR it is expected to remove the COD as well as nitrate nitrogen in the anaerobic phase while in the subsequent aerobic phase ammonia nitrogen will be converted to nitrate nitrogen and phosphorus uptake will be there. In a single cycle the ASBR is capable of removing COD, ammonia nitrogen, nitrate nitrogen and phosphorus.



A-Anode, C-Cathode, MB-Magnetic Bit, RV-Reactor Vessel

Fig. 1. Schematic diagram of electrocoagulation reactor setup

Parameters	Instruments	Methods	
COD	Hatch Reactor	Closed reflux Method	
BOD	_	Modified Winklers method	
Nitrite-Nitrogen	Spectrophotometer	Phenol Di Sulphonic acid method	
Nitrate – Nitrogen	Spectrophotometer	Colorimetric method	
Ammonia –Nitrogen	Spectrophotometer	Nessler method	
pH	pH meter		
Total dissolved solids		Gravimetric method	
Turbidity	Nephelo Turbidimeter	Nephelometeric method	
Chlorides	_	Argentomeric method	
Phosphate	Spectrophotometer	Colorimetric method	
Iron	Spectrophotometer	Phenanthroline method	
Total Hardness	_	EDTA titrimetric method	
Calcium Hardness	_	EDTA titrimetric method	
Magnesium Hardness	_	EDTA titrimetric method	
Calcium	_	EDTA titrimetric method	
Magnesium	_	EDTA titrimetric method	
Heavy metals	Inductively Coupled Plasma	Emission Method	

Table 2. Methodology adopted	for measuring	various	parameter
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The ASBR system was seeded initially with 300 ml of fresh cow dung slurry sieved through 2 mm standard sieve, further it was diluted to 1000 ml with the tap water and was fed with coffee pulping wastewater of 500 ml. It is reported that normally seed culture contains heterotrophs, nitrifiers, denitrifiers and does not contain phosphorus accumulating organisms, hence in order to stimulate the growth of PAOs, the ASBR was fed with coffee pulping wastewater and anaerobic followed by aerobic phase was Initially after seeding was done the provided. biomass required for degradation was developed in ASBR by providing continuous aeration in order to enhance the growth of microbes so has to form the dense culture. After acclimatization period of three weeks, the ASBR cycle was given and operated for a period of 126 days.

From the above tabulation (Table 3), it is clear that, upon electrocoagulation treatment, the parameters concentration of like COD, phosphorus, nitrate nitrogen, and ammonium nitrogen have been reduced to greater extent. Fig 2 (a), (b), (c) and (d), shows the COD, nitrate nitrogen, ammonia nitrogen and phosphorus concentration reduction with days in ASBR. The raw wastewater with the concentration of 24.2 mg/L of nitrate nitrogen, ammonia nitrogen of 60 mg/L and 22.6 mg/L of phosphorus was subjected to the electrocoagulation treatment where the concentration was reduced to 2.6 mg/L, 6.4 mg/L and 2.3 mg/L for nitrate nitrogen, ammonia nitrogen and phosphorus respectively. When this wastewater is subjected to ASBR where on the day 1 nitrate nitrogen was reduced

0.14 mg/L, phosphorus to 1.3mg/L and from day 2 it was BDL. But in case of ammonia nitrogen the concentration was reduced gradually on the day 1 it was reduced to 6.2 mg/L, day 2 it was 6 mg/L, and in day 3 and 4 it was 4.6 mg/L and 2.6 mg/L respectively, but on the day 5 it was 1.2 mg/L and later it was BDL.

3.2 Track Study

Track study was done during electrocoagulation treatment to know the variations in pollutant parameters. Track study was done when raw coffee pulping wastewater was fed to the ASBR. Samples were drawn from the ASBR every 7 minutes time interval. From the Fig. 2 it was observed that COD of the influent sample gradually decreases with increase in time where the initial concentration of COD was 1024 mg/L and it was gradually decreased to 896 mg/L in first 7 minutes, in 35 minutes it was deceased by 84 % of initial concentration. The maximum removal efficiency of COD was 30 mg/L with percentage removal of 97 % which occurred at electrolysis time of 42 minutes.

Fig. 3 and 4 shows the removal of nitrate nitrogen, ammonia nitrogen and phosphorus with time. In the first 7 minutes the nitrate nitrogen was reduced from 5.8 mg/L to 0.87 mg/L, ammonia nitrogen reduced from 15.1 mg/L to 6.06 mg/L and phosphorus was reduce 6.2 mg/L to 1.6 mg/L. By 21st minute nitrate nitrogen, ammonia nitrogen and phosphorus were reduced to 0.58 mg/L, 1.55 mg/L and 0.58 mg/L respectively. Fig.4, specifically gave the best

removal percentage of phosphorus as 96 %, while removal of nitrate nitrogen and ammonia nitrogen was achieved 93 % and 96 %

respectively. When the reaction time was increased the nitrate nitrogen, ammonia nitrogen and phosphorus was BDL.

Parameter	Raw Wastewater	Electrocoagulation Treated Wastewater	Final Treated Wastewater in Anaerobic SBR
рН	4.56	5.04	7.04
Color	Black	Colorless	Colorless
BOD, mg/L	152	276	113
COD, mg/L	3872	768	240
Sulphate, mg/L	4.42	2.26	1.53
Chlorides, mg/L	63.9	31.9	23.9
Phosphorus, mg/L	27	4.36	BDL
Nitrate nitrogen, mg/L	24.2	2.33	BDL
Ammonia nitrogen, mg/L	60	6.3	BDL
Total hardness, mg/L	808	116	240
Calcium hardness, mg/L	60	76	144
Magnesium hardness, mg/L	748	40	96
Alkalinity, mg/L	100	192	136
Aluminum, mg/L	0.91	4.73	1.15
Iron, mg/L	2.49	0.53	0.13
Cadmium , mg/L	0.21	0.21	0.21
Chromium, mg/L	0.26	0.26	0.25
Copper , mg/L	0.17	0.37	0.20
Manganese , mg/L	0.67	0.43	0.41
Nickel , mg/L	0.42	0.24	0.23
Lead , mg/L	0.22	0.20	0.21
Zinc mg/L	0.53	0.14	0.16

Table 3. Performance of Electrocoagulation followed by biological reactor

*BDL-Below Detection Level



Fig. 2. (a), (b), (c), (d). Variation of COD, nitrate nitrogen, ammonia nitrogen and phosphorus in ASBR (RAW)



Fig. 3. Variations of COD during Track Study (7 minutes sampling interval)



Fig. 4. Variation of phosphorus, nitrogen during and ammonia nitrogen during Track Study (7 minutes sampling interval)

During this track study the raw coffee pulping wastewater was added to electrocoagulation reactor and the samples were taken from the reactor at every 15 minutes. The results of this study are shown in Fig. 3 and 4. There was rapid reduction of COD achieved in initial 15 minutes (from 3872 mg/L to 2048 mg/L) and then on the removal was gradual. At the end of 120 minutes the COD concentration was 512 mg/L (83 % COD removal efficiency).

From Fig. 4, it is observed that there was a rapid reduction in concentrations of nitrate nitrogen,

ammonia nitrogen and phosphorus in the first 15 minutes. The nitrate nitrogen concentration reduced from 24.2 mg/L to 6.05 mg/L, ammonia nitrogen reduced from 64 mg/L to 23.8 mg/L while phosphorus concentration reduced from 22.6 to 15.9 mg/L, in first 15 minutes. After 60th minute, the nitrate nitrogen, ammonia nitrogen and phosphorus concentration were, 2.4 mg/L, 12 mg/L and 6.1 mg/L respectively. After 60 minutes the percent removal of nitrate nitrogen and ammonia nitrogen removal was 89 % and 90 % respectively, and phosphorus was 87 %. When the reaction time was increased to 120



Fig. 5. Variations of ICE, Power Consumed during Track Study (16 V)

minutes the concentration of nitrate nitrogen, ammonia nitrogen and phosphorus, reduced to, 2.6 mg/L, 6.4 mg/L and 2.3 mg/L respectively. A comparison of results obtained by electrocoagulation reactor operated at 16 V followed by ASBR is as shown in Table 3. It is observed that when the raw coffee pulping wastewater is treated at 16 V, the removal is appreciable. When the raw efficiency wastewater is treated at 16 V the removal efficiency of COD, phosphorus and nitrate nitrogen was found to be 81 %, 84 % and 90 % respectively.

3.3 Instantaneous Current Efficiency (ICE)

Instantaneous current efficiency (ICE) is the current efficiency measured at constant time interval during the electrocoagulation treatment of wastewater. Measurement of ICE gives information about the formation of polymeric product at the anode during treatment. Melanoidin, a polymeric material present in the coffee pulping wastewater may be adsorbed on the electrode surface and may decrease the efficiency of the electrode. ICE is defined as the ratio of the current stoichiometrically required for the oxidation of organics to that of total consumption. It was calculated using the relation.

ICE (%) =
$$\left\{\frac{(\text{COD})_t - (\text{COD})_{t+\Delta t}}{8 \times I \times \Delta t}\right\} F \times V \times 100$$

Where COD_t and $COD_{(t+\Delta t)}$ are the COD values (gram O_2 per liter) at time t and t+ Δt (seconds) respectively, I is the current in amperes, F is the Faraday's constant (96487 C mol⁻¹), V is the volume of electrolyte (liter), t is the time (seconds) and the value 8 is a dimensional factor for oxygen equivalent mass (g eq⁻¹).

3.4 Determination of Instantaneous Current Efficiency (ICE)

ICE was determined for the raw coffee pulping wastewater for the track study carried out when the electrocoagulation treatment was given at 16 V current. Calculated ICE values were plotted with respect to time (Fig. 5). A common trend has been observed among the plots, the ICE values increased sharply during first 0.5 hour and drop rapidly during further course of electrolysis. Similar trend has been observed by many previous researchers [7,8]. The probable reason for the decrease of ICE may be attributed to the growth of an adherent passivating film on anode surface that might have poisoned the electrode or by production of stable intermediates that cannot be further oxidized by direct electrolysis. possible explanation given by Another Manisankar et al., [8] is that the decrease of ICE to be attributed to the adsorption of melanoidin, a polymeric material present in the wastewater, on the electrode surface or due to the passivation of electrode surface by reaction between the metallic chloride and calcium or magnesium salts present in wastewater. Fig.5 shows the variation of calculated power consumption values with time at 16 V. From Fig.5, it is observed that there was a gradual increase in consumption of power from 0 to 1 hour and sudden increase of power consumption can be seen at 1.5 hour later on there was decrease of power consumption from 1.5 to 2 hour.

4. CONCLUSION

- In the present study aluminum electrode were selected based on the cost effective and easy availability, based on knowledge gathered from the previous studies.
- By using aluminum electrodes, the electrocoagulation treatment resulted in the BOD to COD ratios greater than 0.5 were obtained indicating the increase in biodegradability of the wastewater. Since Coffee pulping wastewater is a high strenath wastewater and after electrocoagulation treatment the COD concentration reduced to 512 mg/L and other parameter such as nitrate nitrogen and phosphorus was reduced to nearly BDL. The COD, ammonia nitrogen, nitrate nitrogen and phosphorus removal were found to be 93 %; 89 %; 90.36 % and 83.85 % respectively. Consequently, it can be concluded that the electrocoagulation treatment method is a promising process for the treatment of coffee pulping wastewater.
- Coffee pulping wastewater is characterized by COD in the range 3584 mg/L to 4980 mg/L, nitrate nitrogen in the range of 21 mg/L to 24 mg/L, phosphorus in the range 22 to 24 mg/L but with low pH which varying between 4.56 to 5.04 for selected source.
- Electrocoagulation treatment with aluminum electrodes operated for 90 minutes followed by biological treatment in anaerobic sequencing reactor operated for 150 minutes will bring down the coffee pulping wastewater characteristics to well below the dischargable standards.
- Track study conducted to know the optimum biological reaction time in ASBR resulted in reduction of COD value from

1024 mg/L to 30 mg/L and BOD from 276 mg/L to 113 mg/L respectively. Nearly complete removal of other nutrients such as ammonia nitrogen, nitrate nitrogen and phosphorous was observed.

 The ICE was determined for the track study of raw coffee pulping wastewater and it was observed that the common trends was followed in all the plots with different current voltages and there was increase in ICE during first 1.5 hour followed by sudden drop for further course of electrolysis.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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