IOSR Journal of Environmental Science, Toxicology and Food Technology (IOSR-JESTFT)
e-ISSN: 2319-2402,p- ISSN: 2319-2399.Volume 9, Issue 9 Ver. II (Sep. 2015), PP 74-82
www.iosrjournals.org

Evaluation of Electrochemical Process for Treating Coffee Processing Wastewater using Aluminum Electrodes

Asha. G [1] and B. Manoj Kumar [2]
[1]*Department of Civil Engineering, Maharaja Institute of Technology, Mysore, Karnataka, India.*
[2]*Department of Environmental Engineering, Sri Jayachamarajendra College of Engineering, Mysore-570006 Karnataka, India.*

Abstract : *Coffee processing is one of the prime agro based industry in India that uses large volume of water with subsequent generation of wastewater. Due to the recalcitrant nature of the compounds in the effluents, disposal is a challenge to the environment. In the present study for comparison, two different set of samples were collected belonging to two different family i.e, Arabica and Robusta. The effluent which is characterized by low pH (3.92 - 4.99 and 4.56 – 5.04) contains high organic load in terms of Chemical Oxygen Demand (COD) (8320 -12840 mg/L and 15360 - 26240 mg/L) and Biochemical Oxygen Demand (BOD_5) (940-1500 mg/L and 3000 – 4200 mg/L), nutrients like ammonia nitrogen (20 – 34 mg/L and 44 – 66 mg/L), nitrate nitrogen (25-30 mg/L and 32 – 45 mg/L), phosphorus (34 – 42 mg/L and 45 – 50 mg/L) etc. The BOD_5 / COD ratio of the sample indicates the less biodegradability of the effluent which indicate load on the receiving body. An effort has been made in this study to compare the treatability of coffee processing wastewater using electrochemical method using sacrificial anode and cathode made of aluminum electrodes to bring down the strength of the coffee processing wastewater into the disposal standards. The effluent concentration of COD reduced to 618 mg/L, ammonia nitrogen reduced to 3.23 mg/L, nitrate nitrogen reduced to 2.4 mg/L and phosphorus concentration reduced to 2.3 mg/L while in case of Robusta, since the effluent concentration of COD and nutrients were high hence was diluted with tap water (1:2) and after dilution the effluent COD, ammonia nitrogen, nitrate nitrogen and phosphorus concentration was 317 mg/L, 1.27 mg/L, 1.12 mg/L and 2.2 mg/L respectively.*

Keywords: *Coffee, Recalcitrant, Electrochemical, Aluminum, Electrodes*

I. Introduction

India is one of the leading producers of good quality coffee for decades, which is grown in southern states of India like Karnataka, Tamilnadu and Kerala[1]. Coffee processing is the prime agro based industry that utilizes large quantity of water. Characterized by high organic load in terms of COD and BOD_5. It also contains high total dissolved solids, total suspended solids, and nutrients like nitrogen, phosphorus, potassium, calcium, magnesium and sulphur. Coffee wastewater is also rich in sugars and pectin's [2], caffeine, fat and peptic substances, macromolecules like lignin's, tannin's, humic acid[3], alkaloid's and polyphenolic's[4]. The final disposal of the pulp and wastewaters containing mucilage and pulp extracts generated by the coffee agro industry is an increasing environmental problem[4],[5] due to recalcitrant nature of the compounds in the effluents. Recently, attention has turned to the electrochemical 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 decolourization treatment and maximum reduction of COD. Electrochemical process is a promising treatment method due to its high effectiveness, lower maintenance cost, less need for labor and rapid achievement of results[6]. With various differences and opinions research in the past few decades have shown that the electrocoagulation is a promising treatment method and effectively potential to treat various types of wastewaters including dye wastewater, tannery wastewater[7],[8], bulk drug manufacturing[9], restaurant wastewater, palm oil mill effluent, food wastewater, potato chip manufacturing wastewater, urban wastewater and removing heavy metals[10], Sugar Wastewater[11], cutting oil emulsions[12], biodigester effluent[13], olive mill wastewater[14,[15].

II. Materials and Methods

1.1. Wastewater Source and Analytical methods

One set of sample belonging to species Arabica was collected from pulping units of coffee estates located in and around Chikkamagaluru and Mercara districts. Another set of coffee processing wastewater belonging to species Robusta was collected from Tata Estate. The samples collected were preserved at 4˚C in a refrigerator for analysis. Samples collected were analyzed before and after treatment for various parameters of concern like COD, BOD_5, ammonia nitrogen, nitrate nitrogen and phosphorus. The samples were filtered through filter paper before analysis. The procedures followed to analyze the parameters of concern were according to the Standard Methods[16],[17]. COD was measured by closed reflux method using Hach apparatus. BOD_5 is measured in terms of Dissolved Oxygen (DO) determination by modified Winkler's method, ammonia nitrogen was measured by Nessler's method by measuring absorbance at 410 nm. In the UV screening method for estimating nitrate nitrogen, UV visible recording spectrophotometer was used for measuring the absorbance at 410 nm. The measurement of UV absorption at 410 nm enables determination of nitrate nitrogen, while phosphorus was measured by vanadomolybdo phosphoric acid colorimetric method by measuring absorbance at 470 nm.

1.2. Batch Experiments

The schematic arrangement of the experimental set up is shown in Fig. 1. The study was conducted in a batch mode electrochemical reactor of 1 L, with a pair of rectangular shaped electrode material made of aluminium[17],[18],[19],[20],[21],[22],[23] with a total area of 49 cm^2, electrodes were connected vertically with an adjustable gap of 1cm between electrodes connected to a DC power supply passing 16V current in monopolar mode. Electrodes were rinsed and washed thoroughly for about 15 min before and after each run with H_2SO_4 (5 % v/v) and water. To maintain homogenity in the reactor magnetic bids were placed. After the optimized electrolysis duration, the system was left undisturbed for about 15-20 minutes and supernatant was drawn for the analysis.

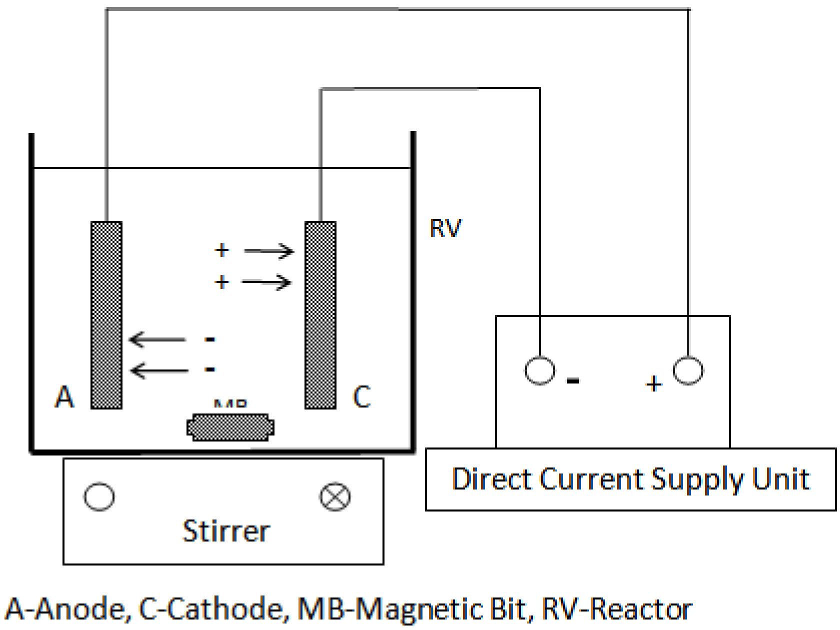

Figure 1: Schematic diagram of electrochemical reactor setup.

III. Results and Discussion

The characteristics results of two different species of coffee processing wastewater collected from two different sources are shown in Table. 1. The coffee processing wastewater belonging to Arabica species was collected from Chikkamagaluru and Mercara estates, Karnataka, India, where as that from Robusta species was collected from Tata Coffee estate. The wastewater is characterized by low pH, high COD, BOD_5, total solids, dissolved solids, ammonia nitrogen, nitrate nitrogen, and phosphorus. The influent concentration of COD for Arabica is 8320 - 12840 mg/L, while that for Robusta is 15360 - 26240 mg/L. Based on the review of comparison and differences from various researchers, aluminum electrodes are used since it is more economical and appreciable for COD and color removal when compared to other electrodes such as stainless steel, copper, iron, titanium etc.

Table. 1 Characteristic of Raw Coffee Pulping Wastewater

Parameter	Concentration	
	Arabica	Robusta
pH	3.92-4.99	4.56 – 5.04
COD, mg/L	8320 -12840	15360 - 26240
BOD_5, mg/L	940-1500	3000 – 4200
Total Solids, mg/L	5000	4758
Total Dissolved Solids, mg/L	4500	4208
Volatile Solids, mg/L	1550	1188
Ammonia -Nitrogen, mg/L	50-84	120 to 180
Nitrate -Nitrogen, mg/L	32-52	135 to 163
Phosphorous, mg/L	60-94	688 to 800
Chlorides, mg/L	63-72	69 – 99
Sulphates, mg/L	4.4-4.5	4-8
Conductivity, µmho/cm	382-390	382.6 – 390.6
Alkalinity, mg/L	85-95	90 – 100
Total Hardness, mg/L	560-750	801 – 808
Calcium Hardness, mg/L	240-350	60 – 63
Magnesium Hardness, mg/L	320-700	720 – 748
Aluminum, mg/L	1.2-1.3	1.26 – 1.37
Cadmium, mg/L	0.15-0.22	0.18 – 0.21
Chromium, mg/L	0.2-0.26	0.20 – 0.26
Copper, mg/L	0.15-0.18	0.17 – 0.18
Iron, mg/L	6.2-6.86	6.3 – 6.83
Manganese, mg/L	0.62-0.67	0.63 – 0.67
Nickel, mg/L	0.42-0.45	0.42 – 0.45
Lead, mg/L	0.22-0.27	0.22 – 0.27
Zinc, mg/L	0.53-0.54	0.53 – 0.54

Initially for Arabica, (Fig. 1) for the first 15 minutes there was no much significant changes in the physical characteristics of wastewater sample, probably evolution of hydrogen at the cathode was not initiated yet, COD removal rate was less than 30 %, later after 30 minutes, as identified by [10] in their work, the COD removal efficiency of 71 % was observed which may be attributed to the fact that the electrode generate coagulated species and metal hydroxides that destabilize and aggregate the suspended particles and precipitate and the hydrogen gas released from cathode would help to float the flocculated particles out of wastewater. During the course of treatment, change in the pH value was noted from 4.56 to 6.22 as a function of time, [24],[25],[26] explained that pH is an important factor that could influence in the overall reactor performance. Further after the electrolysis of 60 minutes, the removal rate of COD was nearing to 90 % indicating higher removal rate with increase in electrolysis duration. Later drastic reduction in COD was observed up to 93 %, i.e. 12840 mg/L to 950 mg/L in 75 minutes with simultaneous increase in BOD_5 value up to 1450 mg/L indicating the biodegradability of the wastewater.

While that in case of Robusta, (Fig. 2) wastewater was diluted since there was no changes in physical characteristics, the reason may be ratio of organic load in terms of COD was more in Arabica compared to Robusta (1:2). Hence the wastewater was decided to dilute with tap water and then initial COD was 6840 mg/L. There was a significant change in COD (43 %) and color removal after 15 minutes. As compared to the removal rate of Arabica, in case of Robusta it was less and noticeable thick color was observed. Hence after 30 minutes, to enhance treatability, an optimized dose of supporting electrolyte NaCl (1.5 gm) was added to increase the conductivity of the wastewater [26], later after 60 minutes, it started reducing gradually from 6840 mg/L to 880 mg/L corresponding to 87 %, which led to the improvement in the efficiency and decrease in power consumption[12]. Later at 90 minutes, nearly 93 % COD removal along with color reduction was observed; this is in line with the observation made by [19] who quoted that COD concentration could be reduced to more than 95 % using NaCl as electrolyte. It can also be observed that as COD decreased from 6840 mg/L to 348 mg/L and BOD_5 increased from 900 mg/L to 2244 mg/L respectively in 105 minutes of electrolysis duration, this increase in BOD_5 concentration is attributed to the fact that some of the organics have broken down into smaller fragments, which are more biodegradable than parent compounds [27].

From Fig. 4 and Fig. 5, for Arabica, it is observed that there was a rapid reduction in concentrations of ammonia nitrogen, nitrate nitrogen and phosphorus from 15th minute onwards corresponding to COD removal until 60 minutes. Ammonia nitrogen reduced from 34.1 mg/L to 4.24 mg/L, nitrate nitrogen concentration reduced from 28.2 mg/L to 3.2 mg/L and phosphorus concentration reduced from 40.6 mg/L to 7.0 mg/L corresponding to 88 %, 89 % and 83 %, respectively. Later after 60 minutes, there was a drop in the removal of ammonia nitrogen and nitrate nitrogen, while maximum phosphorus removal nearing to 95 % was observed, similar trend of phosphorus removal was observed by [21], these findings are in line with [17], who explained phosphorus removal is more effective using aluminum electrodes cause it can be easily ionized and combined with the phosphate ion forming aluminum phosphate ($AlPO_4$) for precipitation. After 75 minutes there was drop in the removal rate which may be attributed to the fact of exhaustion of electrodes with higher oxidation and may be due to pitting corrosion [23]. From Fig. 2, Fig. 3, Fig. 4 and Fig. 5, it is clear that COD and ammonia are removed simultaneously by indirect oxidation during electrolysis and the removal of ammonia is dominant reaction, similar trend was noticed by [28] and reported that the efficiency of electrochemical oxidation of ammonia increases with chloride ion concentration and electrolysis time. While for Robusta, initially there was no significant removal observed for ammonia nitrogen, nitrate nitrogen and phosphorus, after 15 minutes, a slight change in color with less removal efficiency was noticed. Further, after the addition of optimized quantity of NaCl (1.5 gm) at 30 min, ammonia nitrogen reduced from 66.5 mg/L to 32.4 mg/L, nitrate nitrogen reduced from 32.8 mg/L to 15.6 mg/L and phosphorus reduced from 49.5 mg/L to 29.4 mg/L corresponding to 51 %, 52 % and 41 % respectively. Later after 60 minutes, the removal rate was gradual and was reduced to 6.5 mg/L, 5.4 mg/L and 7.4 mg/L corresponding to 90 %, 84 % and 85 %, respectively.

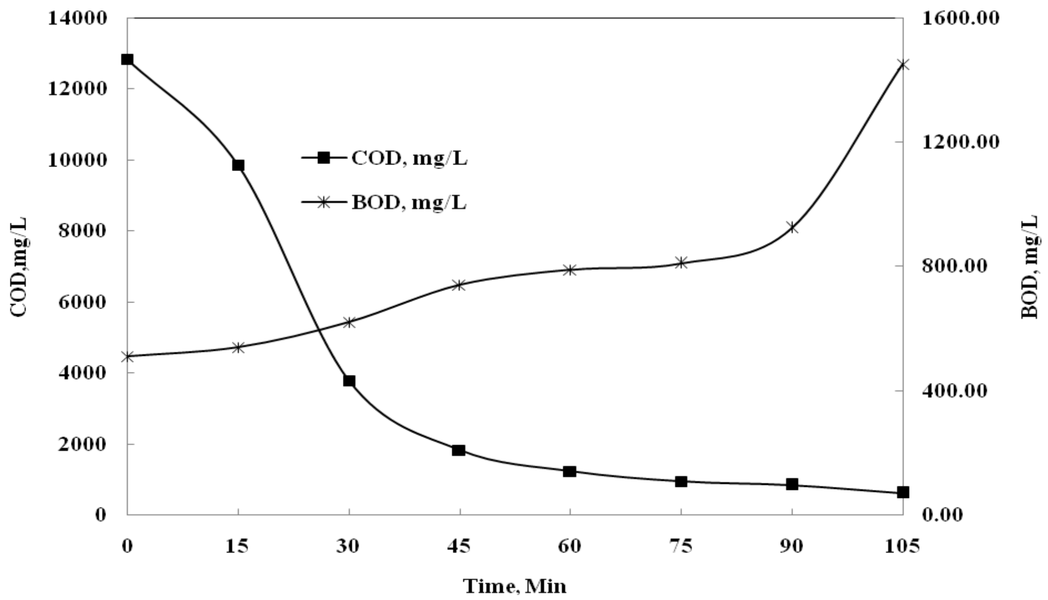

Figure 2: Variation of COD and BOD_5 for Arabica

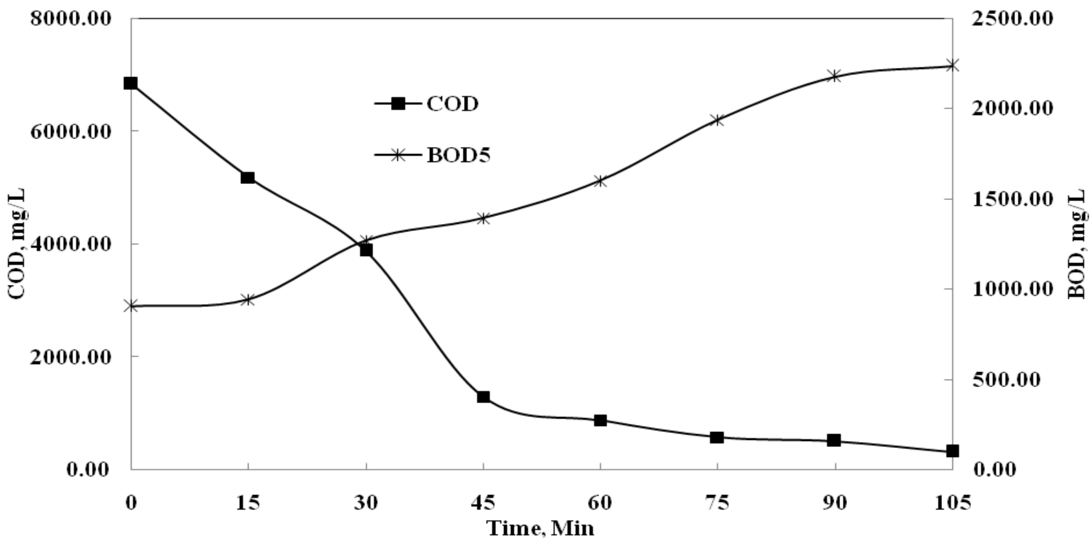

Figure 3: Variation of COD and BOD_5 for Robusta

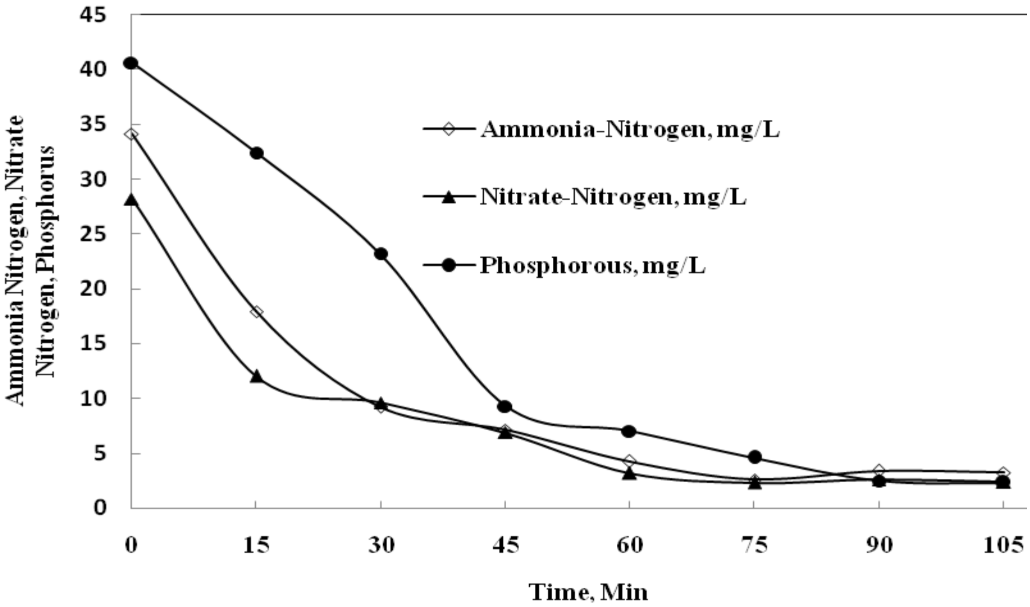

Figure 4: Variation of Ammonia Nitrogen, Nitrate Nitrogen, Phosphorus for Arabica

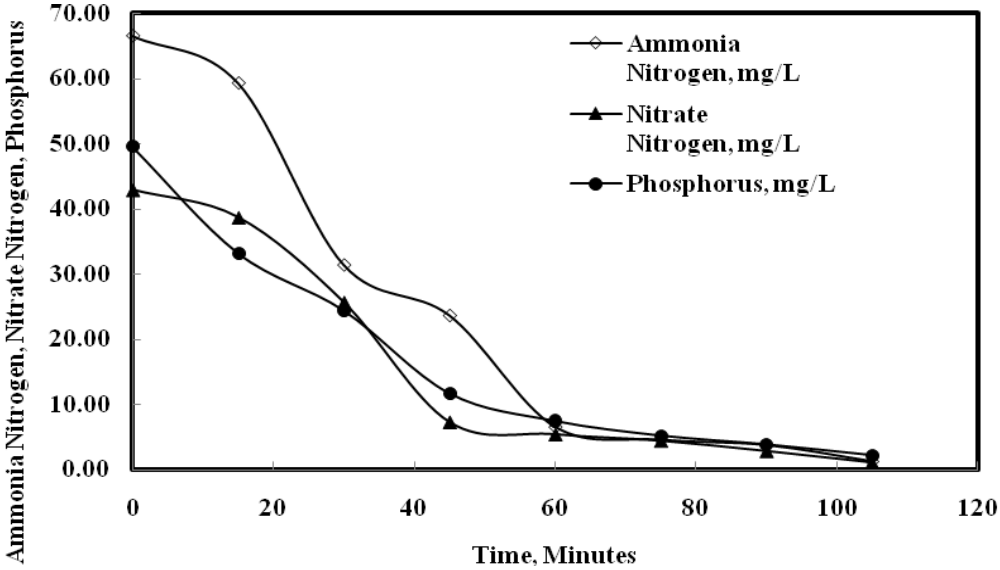

Figure 5: Variation of Ammonia Nitrogen, Nitrate Nitrogen, Phosphorus for Robusta

Instantaneous Current Efficiency (ICE)

The Instantaneous Current Efficiency was determined for the raw coffee processing wastewater for both Arabica and Robusta for electrochemical experiments carried out at 16 V current. Calculated ICE values were plotted with respect to time (Fig. 6 and Fig. 7). A common trend has been observed among the plots, the ICE values of COD, ammonia nitrogen, nitrate nitrogen and phosphorus increased sharply during first 30 minutes and dropped rapidly during further course of electrolysis. It can be observed from Fig. 6 and Fig. 7 that ICE values during the first 30 minutes in case of Arabica are higher varying linearly indicating better degradation during initial electrolysis time. While in case of Robusta, in the first 30 minutes, high values of ICE is only in case of ammonia nitrogen and nitrate nitrogen whereas there was slight drop in ICE value in case of COD and phosphorus with improvement after 45 minutes. A similar trend has been observed by many previous researchers[29],[30],[31]. 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. Another possible explanation given by [30] is that the decrease of ICE to be attributed to the adsorption of melanoidins, a polymeric material [32] present in the wastewater, on the electrode surface or due to the passivating of electrode surface by reaction between the metallic chloride and calcium or magnesium salts present in wastewater.

In case of Robusta, the addition of NaCl lead to the decrease in power consumption because of the increase in conductivity because chloride ions could significantly reduce the adverse effect of other anions such as HCO^{3-}, SO_4^{2-} leading to the precipitation of Ca^{2+} or Mg^{2+} ions[33] forming an insulating layer on the surface of the electrodes which will sharply increase the potential between electrodes resulting in a significant decrease in the current efficiency, this is in line with [25]. The ICE values for Robusta is high compared to Arabica as the strength of Robusta is high which almost twice the strength of Arabica is and hence Robusta was diluted for electrochemical treatment along with the addition of a supporting electrolyte to initiate color removal and load reduction. As observed by [28], In comparison with ICE of ammonia nitrogen in Arabica, it was clearly found that ICE of ammonia nitrogen in Robusta increased with increasing chloride concentration i.e after the addition of NaCl at 30 minutes.

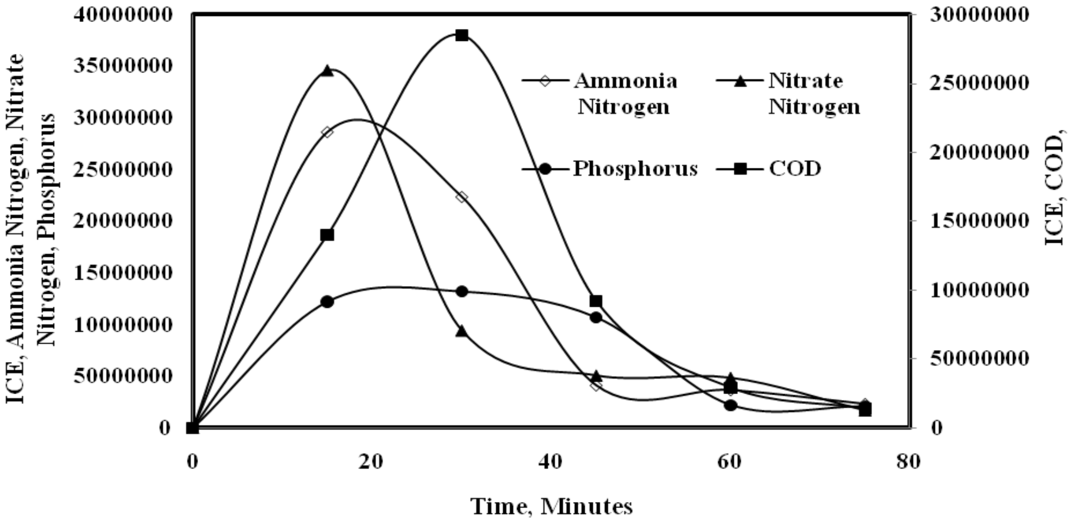

Figure 7: Variation of ICE for Ammonia Nitrogen, Nitrate Nitrogen, Phosphorus for Arabica

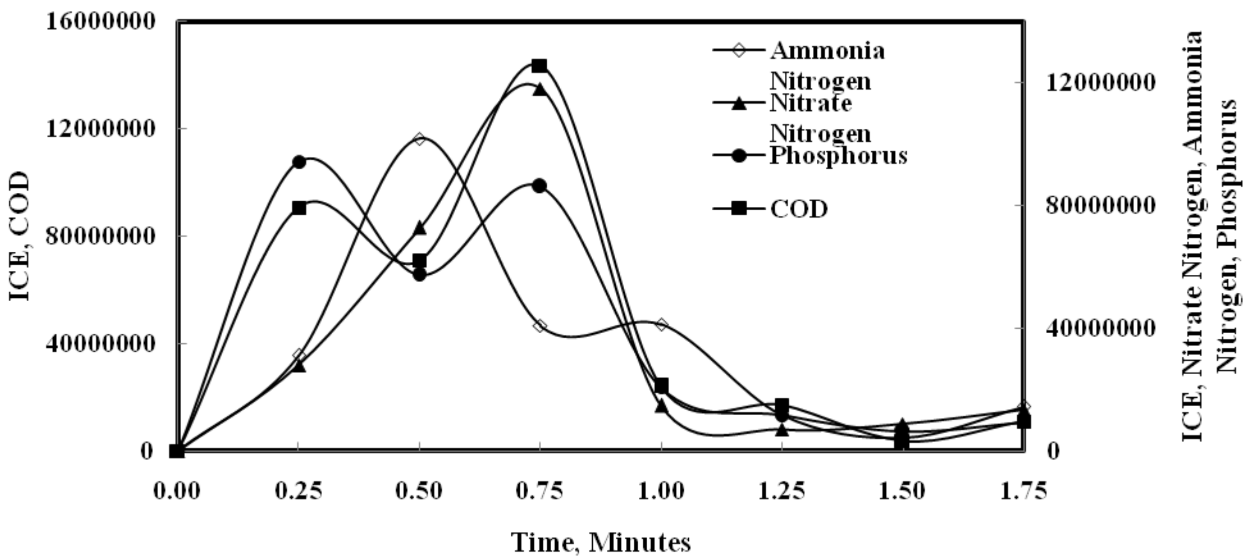

Figure 7: Variation of ICE for Ammonia Nitrogen, Nitrate Nitrogen, Phosphorus for Robusta

Energy Consumption

The energy consumption for the removal of one Kg of COD is calculated and expressed in kWh/Kg removal. The average cell voltage, during electrolysis, is taken for calculating the energy consumption[31]. Fig. 8 and Fig. 9 shows the electrical energy consumption for COD, ammonia nitrogen, nitrate nitrogen and phosphorus. Initially, the energy consumption for pollutant removal in case of Arabica was increasing sharply. These results indicate that the various organic compounds in coffee pulping wastewater have different energy consumption. In case of Arabica, the energy consumed for destruction of COD was high 0.3 to 18.21782 kWh/Kg removal at 90 minutes, followed by nitrate nitrogen, phosphorus and ammonia nitrogen respectively, while in case of Robusta it is 0.6 to 26.6 kWh / Kg, followed by ammonia nitrogen, phosphorus and nitrate nitrogen. For nitrate nitrogen in Arabica it is 0.2 to 29.7 kWh / Kg, while in case of Robusta it is 0.6 to 10.7 kWh / Kg, for ammonia nitrogen in Arabica it is 0.2 to 44.7 kWh / Kg, while in case of Robusta it is 0.7 to 17.1 kWh / Kg, for phosphorus in Arabica, it is 0.7 to 27.9 kWh / Kg, while in case of Robusta it is 0.7 to 11.8 kWh / Kg. Maximum energy demand in Arabica is to treat ammonia nitrogen, while in case of Robusta it is to treat COD. In comparison with Arabica, the energy consumption by pollutants in Robusta was less probably due to the addition of supporting electrolyte, similar trend was noticed by [34] as it is observed from Fig.9.

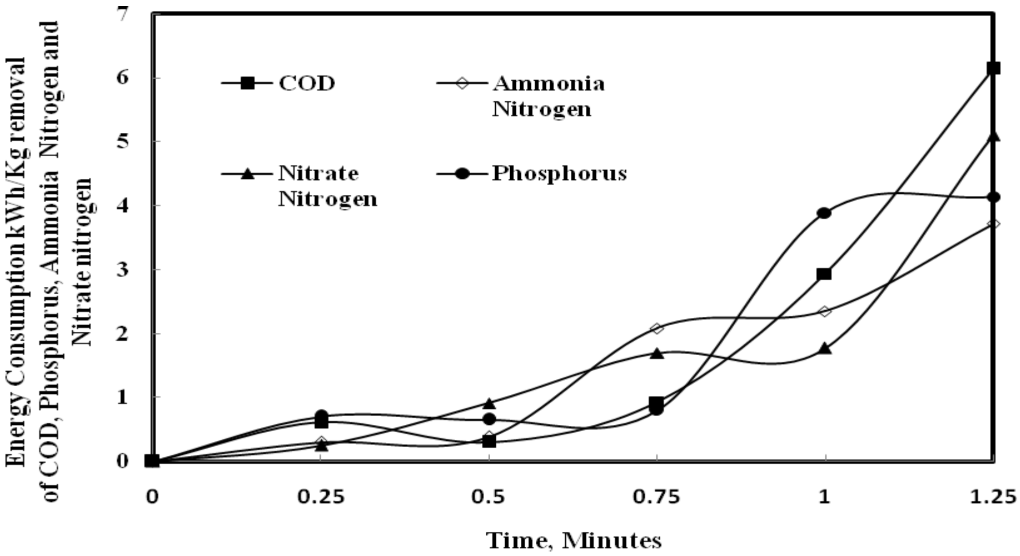

Figure 8: Variation of Energy Consumption for Ammonia Nitrogen, Nitrate Nitrogen, Phosphorus for Arabica

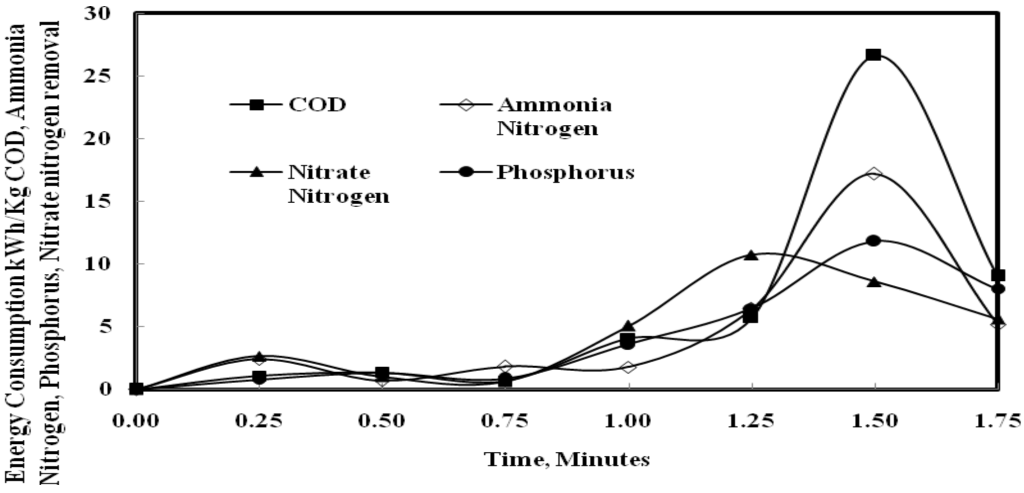

Figure 9: Variation of Energy Consumption for Ammonia Nitrogen, Nitrate Nitrogen, Phosphorus for Robusta

IV. Conclusions

In the present study, attempt has been made to compare the results obtained by treating coffee processing wastewater of two different species i.e Arabica and Robusta. Study revealed that the biodegradability of the recalcitrant wastewater can be enhanced using aluminum electrodes thereby reducing treatment load to dispose safely to the environment. The effluent concentration of COD and nutrients in case of Arabica in the effluent, COD, ammonia nitrogen, nitrate nitrogen and phosphorus concentration was 618 mg/L, 3.23 mg/L, 2.4 mg/L and 2.3 mg/L, respectively. Since the effluent concentration of COD and nutrients in case of Robusta were high it was diluted with tap water (1:2) and after dilution the effluent COD, ammonia nitrogen, nitrate nitrogen and phosphorus concentration was 317 mg/L, 1.27 mg/L, 1.12 mg/L and 2.2 mg/L, respectively. Ratio of organic load is more in Arabica compared to Robusta hence energy consumed for destruction of pollutants in Robusta is high both in COD and ammonia nitrogen and another reason is due to the addition of a supporting electrolyte. ICE and EC was calculated for all the parameters which otherwise was calculated only for COD by previous researchers, since the pollution load was equally important by other parameters identified in the wastewater like ammonia nitrogen, nitrate nitrogen and phosphorus respectively. As observed from previous literature in past few years, the result obtained in the present implies that there is a lower energy requirement for destruction of recalcitrant pollutants.

Acknowledgement

Authors gratefully acknowledges Ministry of Environment and Forests (MOEF), Government of India, for the Financial Assistantship provided for the present work.

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