Reactor design optimization on the electrocoagulation treatment of sugarcane molasses-based distillery wastewater

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Abstract. Electrode geometry and configuration were optimized for the electrocoagulation treatment of sugarcane molasses – based distillery wastewater. Four different reactors, bipolar plates (BP), monopolar plates (MP), bipolar rods (BR), and monopolar rods (MR) were constructed and compared based on their ability to reduce the following wastewater parameters: Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD), Total Suspended Solids (TSS), turbidity, and oil and grease. Other electrocoagulation parameters such as current supplied, applied voltage, aspect ratio, and specific electrode area were held constant. The BP reactor yielded the highest reduction percentage among the parameters: COD = 43\%, BOD = 49\%, TSS = 47\%, turbidity = 54\%, and oil and grease = 79\%. Moreover, the BP reactor gave the least metal consumption at 0.47 kg/m\textsuperscript{3} after 1.5 hours of treatment, thus making it the best reactor design among the four reactors considered.

1. Introduction
Sugar mill factories, which supply molasses to distilleries, discharge large amounts of wastewater with high organic materials, oil, grease, sugar cane juice, syrup and molasses. These materials become environmental pollutants if discharged without treatment [1,2]. Molasses distillery producing wastewater called “spent wash” is considered as one of the most polluting industries due to its effluent, which contains a variety of high molecular weight organics, including caramels, waxes, lignins and melanoidins that defy biological digestion [3,4]. The effluent is characterized by the Central Pollution Control Board (CPCB) in 1994 and 2003 to have enormously high COD (80,000–100,000 mg/l) and BOD (40,000–50,000 mg/l), apart from low pH, high potassium, phosphorus and sulfate content, strong odor and dark brown color [5]. The effluent’s dark color, caused by the presence of melanoidins, is barely affected by biological treatment [6] and hinders photosynthesis by blocking sunlight thereby harming aquatic life [7]. In addition, cane molasses spent wash contains low molecular weight compounds such as acetic acid, lactic acid, ethanol and glycerol [8]. Hence, it is considered to be the most complicated and problematic organic industrial effluents; and appropriate treatment is therefore necessary before the effluent is discharged.

Electrocoagulation (EC) is a modern electrochemical method of treating agro-industry wastewaters [9]. It is a wastewater treatment based on an electrochemical principle, which uses an electrochemical cell having a DC voltage source applied commonly to iron or aluminum electrodes, while the wastewater serves as the electrolyte [10]. Meas \textit{et al} [11] characterized electrocoagulation as a process
involving in situ generation of coagulants by the dissolution of metal from the anode with simultaneous formation of hydroxyl ions and hydrogen gas at the cathode. This process produces the corresponding aluminum or iron hydroxides and/or polyhydroxides, which serve as the adsorbent where pollutants get trapped. The generated gas also helps to float the flocculated particles at the water surface. Electrocoagulation offers many advantages such as in situ coagulant production, simple installation, lower secondary pollution, removal of odor and color, and decreased residence times [12]. Through the years, electrocoagulation has found many applications in various types of wastewater and removal/reduction of wastewater parameters [13-20]. Likewise, it has been used in the treatment of molasses-based distillery wastewater with different considerations such as reactor design [21], electrode material [22,23], coagulant dosing [24], additive [25] and configuration with other technology like microfiltration [26], ozone [27] and electron beam [28] and among others.

This study designed an electrocoagulation system for the treatment of distillery wastewater from a local distillery via a batch-mode process. Specifically, the most appropriate electrode configuration (monopolar and bipolar) and geometry (rectangular plate and cylindrical rods) were investigated for the design of the reactor. Also, possible causes for the differences in the outcomes of electrocoagulation treatments using different reactor designs were determined. The electrode material used was aluminum and other parameters such as current supplied, applied voltage, aspect ratio and the specific electrode area were held constant. The effectiveness of the designs was checked using the following parameters: BOD5, TSS, oil and grease, pH, turbidity and COD. Monitoring of electrode consumption, current efficiency and pH was done to further check and validate the occurrence of the electrocoagulation process in the reactors.

2. Materials and method

2.1. Materials and wastewater

The reactor electrodes were made up of aluminum to serve as sacrificial anodes to continuously produce ions in the water to neutralize the charges of the particles and thus begin the coagulation. Aluminum was chosen for its high stable oxidation state, low solubility, formability and low cost. The electrodes were fabricated at a local metal fabrication shop (Alonzo St., Sta. Cruz, Manila). The wastewater was taken from a local distillery (Sagbat, Nasugbu, Batangas), which utilizes sugarcane molasses in extracting ethanol. The wastewater was taken fresh from the effluent pipe after the biological treatment facilities before discharge to the lagoon. The wastewater was shipped cold immediately from factory to laboratory. The wastewater samples were kept at 4°C during their storage period that lasted from 1-7 days according to Standard Methods (American Public Health Association, 1992). Initial characterization of the distillery wastewater was performed to determine the COD, BOD5, TSS, turbidity, oil & grease, and pH using industrially accepted methods of analysis.

2.2. Experimental set-up

The electrocoagulation process was carried out at a laboratory scale via batch operation. Two different electrode geometries were studied. The batch plate reactor used rectangular plate electrodes while the batch rod reactor employed rod electrodes. Also, two electrode configurations were considered: the monopolar and the bipolar modes. In the monopolar configuration, each electrode was connected to the power supply while in the bipolar mode; only the outmost electrodes were physically connected to the power supply. The electrocoagulation setup is shown in figure 1 while the electrode configurations, monopolar-plates (MP), bipolar-plates (BP), monopolar-rods (MR), and bipolar-rods (BR) are shown in figure 2. The specifications of the reactors and cells are listed in table 1. The reactor operated at normal room conditions under a constant reaction temperature of 25°C and atmospheric pressure. DC power was supplied to the electrolytic cell at constant supplied current settings (1.55 A) and its corresponding voltage settings. Samples were taken at 0.5 h, 1.0 h, and 1.5 h during the study for analysis.
2.3. Reactor design and EC process description

Thirty-six batch runs of electrocoagulation treatment on distillery wastewater were carried out to determine the effects of the electrode configuration and geometry setting on the reduction of wastewater parameters such as COD, BOD5, TSS, turbidity, oil and grease, and pH. The reactor vessel was made from glass due to its inert properties suitable for the EC process. Tall vertical plate and rod electrodes were fabricated based on the dimensions shown in table 1 which make the vessel aspect ratio greater than 5:1 to increase the process efficiency [21]. This reactor shape allowed gas bubbles to flow from bottom to top. The reactor has a reaction zone of 5.3 L with an overflow allowance of 1 L to accommodate the bubbles expected to be generated along the treatment process. The electrodes were
made from aluminum due to its negative standard reduction potential value of \(-1.67\) V, making it readily oxidized than reduced which is suitable for the production of coagulants in the EC process. The electrode dimensions as given in table 1 were fixed such that the area of the plate and rod reactors will have the same value of 0.0458 m\(^2\) and thus eliminate the effect of area on the electrocoagulation process. Four arrangements of aluminum electrodes (figure 2) were examined to evaluate its performance in the electrocoagulation treatment of distillery wastewater. The monopolar arrangement with all four plates connected to a power supply and arranged such that the two anodes and two cathodes were arranged alternately, while the monopolar arrangement with 25 rods placed 5 x 5, consisting of 10 anodes and 15 cathodes arranged alternately were all connected to the power supply. On the other hand, the bipolar arrangement only had the extreme electrodes connected to the supply. It also utilized four plates and the other with 25 rods with the internal plates/rods being bipolarized with opposite charges facing each. The plate electrodes were connected to the power supply using probe wires or clips while the rod electrodes were connected by using a bolt and nut assembly. The spacing of 23 mm for the plate reactors and 10 mm for the rod reactors was a result of the effort to fix and equate the area of the two types of reactors with different geometries. It was the highest possible way to maintain equal spacing in between electrodes and the reactor wall. The current supplied was kept constant at 1.55 A to eliminate the effect of varying current settings on the treatment process. The voltage supplied varied depending on the current settings since the power supply was operated under constant current mode.

Table 1. Electrocoagulation reactor specifications.

| Parameters       | Description                        | Parameters       | Description              |
|------------------|------------------------------------|------------------|--------------------------|
| Vessel           | Borosilicate glass (Pyrex)         | Electrode Material | Aluminum               |
| Material         | Vertical rectangular prism         | Shape            | Rectangular strips       |
| Shape            |                                     | Dimensions       | Cylindrical rods         |
| Dimensions       | Length = 100 mm                    |                  | Strips                  |
|                  | Width = 100 mm                     |                  | Width = 86.5 mm          |
|                  | Depth = 630 mm (530 mm submerged)  |                  | Depth = 630 mm           |
|                  |                                     |                  | Thickness = 3 mm         |
|                  |                                     |                  | Rods                     |
|                  |                                     |                  | Outer Diameter = 11 mm   |
|                  |                                     |                  | Depth = 630 mm           |
| Volume           | Total = 6.3 L                      | Total effective area | 0.0458 m\(^2\)          |
|                  | Reaction zone = 5.3 L = 0.0053 m\(^3\) |                  |                          |
|                  | Overflow allowance = 1 L           |                  |                          |
| Operation        | Batch mode                         | Number           | 4 rectangular strips     |
|                  |                                     |                  | 25 cylindrical rods      |
| Power supply     |                                     | Configuration    | Monopolar, bipolar       |
| Voltage          | Corresponding voltage at current setting |                  | Parallel, equally spaced |
| Current          | 1.55 A                             | Spacing          | Strips Spacing = 23 mm   |
|                  |                                     |                  | Rods Spacing = 10 mm     |
| Optimized parameters | Depth:width = 5:1                | Specific electrode area | Effective area: vessel volume |
| Vessel aspect     |                                     |                  | = 77 m\(^2\)/m\(^3\)   |

2.4. Wastewater analysis
The wastewater samples were tested in accordance to methods used by local industries. For COD, turbidity, and TSS, samples were analyzed using a Hach DR/2010 portable datalogging
spectrophotometer. The sample was contained in the vessel provided by the instrument. Oil and grease content was evaluated by the Hexane Extractable Gravimetric Method using 30 mL hexane in 1 L of sample. The two layers were separated via extraction and the hexane layer was filtered, solvent evaporated and the remaining residue was weighed to obtain the oil and grease content of the sample. BOD\textsubscript{5} was measured using a BOD Merc-free sensor that automatically reads the sample’s BOD in ppm, in which 215 mL of sample was transferred to the BOD bottle together with 215 mL of diluted BOD nutrients. The bottle was stoppered with rubber having a magnetic stirrer inside with 3 pieces of NaOH pellets. The BOD bottle was then closed in the BOD sensor device and was agitated. The BOD\textsubscript{5} was then read from the device after the five-day period. The pH of the wastewater was determined using an OHAUS pH meter (Starter 2000 model). Visual inspection of color was done for qualitative analysis of the wastewater sample while measuring turbidity was used for quantitative analysis.

3. Results and discussion

3.1. Effect on wastewater parameter removal

The reduction of wastewater parameters (COD, BOD\textsubscript{5}, TSS, turbidity, and oil and grease) from the electrocoagulation treatment process was measured to verify the effect of efficiency in using the proposed electrode geometry and configuration. The results of each electrocoagulation treatment per reactor design on wastewater parameter removal at different time interval are presented herein. Figure 3 showed that the treatment using the BP reactor gave the highest removal percentages among all the wastewater parameters considered, followed by the MP, BR and MR reactors. Likewise, among the three time interval considered, the 1.5 h has the highest percentage removal for all the parameters considered. Summarized in table 2 is the result at 1.5 h and will the subject of discussion for all the parameters since it is the highest value obtained.

It can be seen in table 2 that treatment via BP EC reactor resulted in the highest COD removal followed by MP, BR, and then MR. These results correspond to the results obtained by other researches that applied electrocoagulation in the treatment of synthetic textile wastewater [20], slaughterhouse wastewater [16], fluoride contaminated water [18] and aqueous solutions with Cr\textsuperscript{3+} [29] that concluded that bipolar plate electrodes produced higher COD removal efficiencies than monopolar plate electrodes. The bipolar reactors BP and BR have had higher COD removal rates than their monopolar counterparts MP and MR. This behavior could be attributed to the higher current densities present in the bipolar electrodes than in the monopolar ones for the same amount of current supplied which resulted to higher coagulant dosage and thus, resulted to the removal of more pollutants in the wastewater. On the other hand, the plate reactors BP and MP have had higher COD removal rates than their cylindrical rods counterparts BR and MR since current is more uniformly distributed in the former than in the later which resulted to the fewer occurrences of parasitic reactions in the reactor which negatively influences the system by competing with the main reaction of electrocoagulation.

Correspondingly, based from table 2, the same trend with COD was observed wherein BP reactor had the highest BOD\textsubscript{5} removal efficiency, followed by the MP, BR, and MR reactors. This is just realistic because both BOD\textsubscript{5} and COD measures oxygen demand only in a different manner, the former relying on enzymes produced by bacteria to catalyze the oxidation of organic matter during a five-day incubation period while the latter is based on using chemical oxidants to oxidize organic matter. Thus, presumably the higher current densities resulted to a greater enzyme production.

The removal of total suspended solids or TSS was measured as well for all the four reactors and the same trend was observed: BP > MP > BR > MR. The removal of suspended solids increased with time since during the EC treatment, metal ions acted as destabilization agent of finely dispersed particles. Hence, the longer the treatment time, the more particles were destabilized resulting to breaking of emulsions, particulate suspension and eventually aggregation forming flocs [10].
Figure 3. Percent removal of wastewater parameters (COD, BOD5, TSS, Turbidity, Oil & grease) per reactor (BP = bipolar plate, MP = monopolar plate, BR = bipolar rod, MR = monopolar rod after a) 0.5h, b) 1.0h, and c) 1.5 h of electrocoagulation treatment.

Table 2. Percent wastewater parameters removal at 1.5 h using various electrode geometry and configurations.

| Parameters          | BP         | MP          | BR          | MR          |
|---------------------|------------|-------------|-------------|-------------|
| COD (ppm)           | 43.10 ± 0.0038 | 33.65 ± 0.0042 | 15.97 ± 0.0061 | 11.15 ± 0.0066 |
| BOD5 (ppm)          | 49.06 ± 0.0170 | 36.32 ± 0.0170 | 18.69 ± 0.0169 | 13.79 ± 0.0170 |
| TSS (ppm)           | 47.14 ± 0.0044 | 36.56 ± 0.0044 | 32.16 ± 0.0044 | 20.85 ± 0.0135 |
| Turbidity (FAU)     | 54.27 ± 0.0110 | 34.15 ± 0.0081 | 30.59 ± 0.0093 | 28.05 ± 0.0092 |
| Oil & Grease (g/L)  | 78.65 ± 0.0117 | 73.41 ± 0.0056 | 64.79 ± 0.0067 | 55.80 ± 0.0082 |
Water turbidity could be reduced by electrocoagulation [15] and similar trend was observed for turbidity: BP > MP > BR > MR. Turbidity is related to TSS, since reduction of turbidity resulted from the destabilization of colloids due to the effect of the electric field generated between the electrodes and the reactions with coagulating compounds formed in situ during anode oxidation, followed by a subsequent flotation of agglomerates of the particles [30]. Thus, the more particles are destabilized as in TSS, the less is the turbidity. This could easily be associated with the color change of the wastewater as a function of time as shown in figure 4. The wastewater color changes from very dark brown to a lighter color especially from using bipolar plate electrode.

![Figure 4. Wastewater color change using (a) bipolar plates reactor, (b) monopolar plates reactor, (c) bipolar rods reactor, and (d) monopolar rods reactor.](image)

Furthermore, the trend for oil and grease removal is the same: BP > MP > BR > MR. This observation is similar with the findings of Phalakornkule et al [31] in 2010 where an increase in current density, opting for bipolar than monopolar [16] and plate over rod [32] will result in an increase of oil and grease removal. Oil and grease removal as a function of charge loading is mainly attributed to the size of gas bubbles whose specific area decreases with the diameter of gas bubbles produced during electrolysis. An increase in current density will increase the specific area of the gas
bubbles which means that there will be also an increase in small colloidal particles attached to it. Hence, the large gas bubbles can interfere with the oil-removal process by reducing the probability of oil droplet collisions and the probability of attachment of oil droplets to flocks, thus reducing the oil and grease removal efficiency.

Using ANOVA: two way factor without replication statistical analysis at 95% confidence level, the reactor type, electrode configuration, and treatment time were found to individually both have significant effects on all wastewater parameters considered (COD, BOD5, TSS, turbidity, and oil and grease) since $F > F_{critical}$ and $P$-value $< \alpha$. Thus, these design parameters should be considered in the construction of reactors and electrodes for wastewater treatment using electrocoagulation.

3.2. Reactor design

The efficiency and end products of electrochemical waste treatment are profoundly dependent on the characteristics of the electrodes used such as material type, configuration, and geometry [33]. Bipolar reactors experience higher current densities with 33 A/m² (BP and BR) than monopolar reactors with 11.27 A/m² (MP) and 8.46 A/m² (MR) from the same amount of supplied current (1.55 A) due to their electrical configuration of being in series that resulted into equal amounts of current flowing through the electrodes as compared to the monopolar reactors that are in parallel connection that resulted in divided amounts of current. This explains why the bipolar reactors performed better than the monopolar reactors. On the other hand, electrode geometry dictates the current distribution profiles of electrodes which is one of the key factors in the design of energy-efficient electrocoagulation reactors [32]. It highly influences the electrocoagulation due to the current distribution on the surface of two electrodes and its effect on energy utilization. Plate reactors have uniform current distribution represented by straight current lines along its surface in between the edges and non-uniform current distribution or dissipation of current lines as represent by curved current lines on its edges due to the increase in electric field in this region caused by the current flow that passes partially around the rectangular space filled with wastewater [34]. On the other hand, rod reactors have non-uniform current distribution all throughout its surface. The non-uniformity or non-homogeneity of the current distribution patterns is indicative of wasteful increases in energy releases along the edges of the plates and surface of the rod since this increase in local current density is believed to be utilized by the parasitic reaction of water electrolysis that generates unwanted oxygen and hydrogen gas in the cell [9,16,29,32,35-37]. This explains why rectangular plate reactors performed better than cylindrical rod reactors.

3.3. Effect of operational parameters in EC

Operational parameters such as pH, electrode consumption, and current density should also be considered during the electrocoagulation process to account for its occurrence and efficiency of the process. As shown in figure 5, the pH of wastewater increased in all types of reactor used as the time of treatment increased due to the accumulation of $\cdot OH$ ions during the hydrolysis of metal producing metal hydroxides and poly hydroxides [10]. This result conform to the findings of Chen [17] and Kobyta [38] that pH increases during the process.

The electrode consumption for the electrocoagulation treatment of distillery wastewater was also investigated to verify the formation of coagulants. The total metal losses obtained by taking the difference between the final and initial masses per electrode were compared for the four types of reactor used. The results shown in figure 6 are reported in terms of the change in mass divided by the volume of wastewater treated to estimate the electrode costs per volume of wastewater. Both bipolar electrodes plate (0.09 kg/m³) and rod (0.13 kg/m³) had less metal consumption than monopolar electrodes plate (0.24 kg/m³) and rod (0.42 kg/m³). Although bipolar reactors were expected to produce more coagulants based on higher electrode consumption due to their higher current densities than the monopolar reactors, the occurrence of the parasitic reaction of water hydrolysis lessened the formation of coagulants [35]. In addition, the less metal consumption means more economical since less electrode is needed to affect the electrocoagulation process.
Another observation made during the experiment was that the actual electrode consumption was lower than the theoretical consumption as predicted by Faraday’s law. This observation is best presented using current efficiency values. In figure 7, BP had the lowest current efficiency with 61% followed by BR, MP, and MR with 65%, 80%, and 84%, respectively. Current efficiency or Faradaic efficiency is the ratio of the actual metal loss over the theoretical loss as predicted by Faraday’s law [29]. The same study observed that bipolar systems have shown lower current efficiencies than monopolar systems. The actual metal loss is different from the theoretical loss since Faraday’s law assumes that all the current supplied is passed thru the cell and does not consider other factors that may affect metal dissolution such as the actual amount of current that passes thru the cell which, in this case, is affected by the electrode configuration and geometry as discussed in the previous section regarding the differences in organics removal in different electrocoagulation reactor designs. The current efficiency was observed to decrease with treatment time (electrode usage). This observation may be attributed to the visually observed increase in surface roughness with treatment time. Roughness leads to dramatic reduction of the electrical conductivity of metals due to the inter sheet electron scattering at the Fermi surface which is perturbed by roughness, thus disturbing the smooth flow of electrons which is the principal mechanism of electrical conduction [39]. The increase in surface roughness with time and thus the decrease in current efficiency over time only support the
validation of the occurrence of electrocoagulation process in the cell since the electrodes were proven to experience surface corrosion. The current efficiencies may be used in the estimation of operational costs in future applications.

![Current efficiency profile of different reactor as a function of time.](image)

**Figure 7.** Current efficiency profile of different reactor as a function of time.

4. Conclusion
Distillery wastewater contains high organic contents that pose serious environmental problems, thus requiring a highly efficient wastewater treatment process. Electrocoagulation was proven to be an effective preliminary treatment in reducing wastewater’s COD, BOD, TSS, turbidity, and oil and grease content. Electrode geometry and configuration were optimized for the electrocoagulation treatment of wastewater via a batch mode process using aluminum electrodes. The following conclusions can be drawn from this study: 1) High current in the cathode results in parasitic reactions that disturb organic matter deposition and co-precipitation with metallic hydroxides; thus, bipolar reactors (BP and BR) resulted in higher wastewater parameter reduction efficiencies than their corresponding monopolar counterparts (MP and MR) due to their less scattered current distribution profiles which caused lesser occurrence of parasitic reactions such as water electrolysis. 2) Bipolar plates became the optimum electrode setup due to its inter-electrode spacing, more efficient use of energy, better supply of current, and better current distribution than cylindrical rods reactors in removing high organic matters that constitute wastewater parameters such as COD, and BOD. They also produce less hydrogen bubbles that increase colloidal particles to form flocs and oil attachment to flocs which constitute wastewater parameters such as TSS, turbidity, and oil and grease. Statistical analysis showed that there is a significant relationship between the reactor geometry and configuration and the removal of pollutants. Thus, these design parameters should be considered in future applications of electrocoagulation.

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