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# Electrochemical treatment of Ayurveda pharmaceuticals wastewater: Optimization and characterization of sludge residue

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ABSTRACT

In present study, we have investigated the treatment of Ayurveda pharmaceutical wastewater (APW) by electrocoagulation (EC) treatment method to minimize the energy consumed per unit mass of COD removed and maximize the color and COD removals by using the stainless steel (SS) and aluminum (Al) electrodes. Response surface methodology (RSM) was used to optimize the multiple responses during the batch mode study of APW treatment. A full factorial central composite experimental design has been used to determine the experiments for each factors and optimize the responses: color removal ($R_1$), COD removal ($R_2$), and energy consumed per unit mass of COD removed ($R_3$). At optimized conditions, $R_1$= 97.83%, $R_2$ = 58.35% and $R_3$ = 27.12 kWh/kg COD removed for Al electrodes and $R_1$ = 95.35%, $R_2$ = 78.88% and $R_3$ = 28.32 kWh/kg COD removed for SS electrodes, respectively were obtained. Field emission scanning electron microscopy (FESEM) and thermo-gravimetric analysis (TGA) have been used to determine the EC treatment mechanism and disposal aspects of EC solid residue (scum and sludge). EC treatment mechanism of APW seems to be a combination of electro-floatation, electro-oxidation, and electro-coagulation.



## 1. Introduction

Ayurveda or Ayurveda medicine also called “science of life” or “science of longevity” is one of the oldest medical systems all over the world. More than 3000 and 520 BC ago Ayurveda medicines appeared and developed in India [1,2]. India is one of the oldest country in the world where traditional medicinal health systems are found. In India, 70% population health treatment depends upon the Ayurveda medicine system (AMS) and approximately 40% peoples of western countries using the same medicine system for various diseases treatment [3]. Accordingly, the interest in the use of traditional medicines is rapidly growing. Due to this attention, Indian government and other world institution supports the research of AMS to overcome the side effects, cost factor and adverse drug reactions of the modern medicines. Currently, 250,000 Ayurveda system registered to medical practitioners as compared 700,000 modern medicine in India [4,5]. More than 20,000 medicine plants have been reported while 7100–7500 plants are used by traditional practitioners for curing the different diseases [6]. Traditional medicinal system for health care are used more than 1.5 million practitioners in India. It is predictable that around 7850 manufacturing units are participate in traditional plant-based formulations and in the production of natural health products in India.

It is to be noted that Ayurveda pharmaceuticals manufacturing (APM) units are using a large volume of water during the various step of manufacturing and produce the huge quantity of higher strength polluted wastewater. The APM effluent contains the suspended particles, heavy metal ions, higher pH, strong color, higher chemical oxygen demands (COD) and including a large number of other impurities. Therefore, APM units are one of the leading manufacturing units generating a higher strength and huge volume of highly concentrated polluted wastewater. APM effluent can severe rigorous pollution if not imply proper pretreated processes before mixed in conventional wastewater treatment plants (WWTPs) and/or discharge to the water bodies.

A large number of treatment techniques include membrane filtration, coagulation–flocculation, ozonation, adsorption and advanced oxidation processes (AOPs) have been proposed for the treatment of such kinds of highly concentrated wastewater [7–15]. However, APW contains the hydrophilic nature with a low adsorption rate and higher biological activity even in very

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low concentration. Hence, APW is highly persist for biological pretreatment [9–12].

Recently, electrochemical treatment technology (ECT) including electrocoagulation, electroflotation, electrodissolution, electroflocculation and electrooxidation is a most convenient and effective technique for the treatment of highly concentrated wastewaters. Various authors have been investigated the applicability of ECT for various type of industrial wastewaters [7–16]. In electrocoagulation, usually aluminum (Al), iron, and stainless steel (SS) electrodes are used as electrodes and they act as an active coagulant precursors by the anodic dissolution and hydrogen gas evolve on cathode appears as gas bubble which is interact with impurities via the electroflotation resultant impurities accumulated at the surface of the solution during the treatment [17,18].

Numerous local Ayurveda manufacturing units are also producing a large quantity of APW. In most of the cases these effluents are discharged into open channels without proper treatment [1–3]. Hence, a treatment methodology is required which is easy to operate and can be easily accepted by these local Ayurveda manufacturing units. EC technique is one of the possibility for the treatment of such types of highly concentrated wastewaters [19–21].

In present work, the performance of EC process was evaluated by using the four operational process parameters namely, solution pH, electrolysis time (t), current density (j) and electrolytic concentration (m) as input variable during the response surface methodology (RSM) optimization. A full factorial central composite design (CCD) was applied for determine the experimental runs for the three responses namely, color removal, COD removal, and energy consumption per unit mass of COD removed. Desirability function approach for multi-response optimization in terms of maximize the color and COD removal and minimize the energy consumption has also been used in present study. Physicochemical analysis of solid residue generated by Al and SS electrodes has been done to understand the mechanism of EC process. Disposal aspects of residue have been proposed on the base of the thermo-degradation analysis.

## 2. Material and method

All chemicals of analytical grades were used in present work. Sulfuric acid, hydrochloric acid, sodium hydroxide, potassium dichromate, mercuric sulfate and silver sulfate were purchased from Ranbaxy Chemicals Ltd., New Delhi, India.

### 2.1. Experimental setup and methodology

EC experiments were performed in a rectangular lab-scale batch reactor, made of perspex glass (130 mm × 130 mm × 210 mm) having 1.0 l reactor volume. One pair of Al and SS plates electrodes having the thickness (1.5 and 2.5 mm) and dimension (8.5 cm × 9.5 cm) with 82 cm$^2$ effective surface area was used in each experimental run. The electrode gap between the two electrode plate was maintain ~1.0 cm in all experimental runs and electrodes were kept ~5.0 cm above from the bottom of reactor for homogenous agitate the solution by using the magnetic bar and the reactor put on the magnetic stirrer. All experiments were completed at 30 ± 5°C. 0.9 l of wastewater was treated in EC reactor during each experiment run. Hydrochloric acid (0.1 N) and sodium hydroxide (0.1 N) aliquots was used for adjusted the solution pH from initial level to desired value. Sodium chloride used as an electrolyte for adjust the initial conductivity of wastewater. Parallel arrangement mode of the electrodes was used for the present study. Electrodes distance during the study fixed and controlled by using the non-conducting wire. A precision digital direct current power supply (1–15 V, 0–5 A) was used to maintain the constant current density. The APW was collected

**Table 1**
The APW characteristics before treatment and after treatment the solution at the optimum conditions.

| Wastewater Characteristic Parameters | Value | | |
|---|---|---|---|
| | Before treatment | After treatment | |
| | | SS | Al |
| Chemical oxygen demand (mg/l) | 4200 | 78% | 68% |
| Color (Platinum cobalt unit) | 7200 | 97% | 98% |
| Turbidity (NTU) | 180 | 23 | 29 |
| Conductivity (mS/cm) | 5.3 | 3.2 | 3.9 |
| pH | 8.9 | 10.4 | 8.9 |
| Total solids (g/l) | 17.89 | 0.61 | 0.73 |
| Total dissolved solid (g/l) | 13.32 | 0.48 | 0.59 |
| Total suspended solid (g/l) | 3.4 | 0.19 | 0.24 |

from the local Ayurveda manufacturing units located in Haridwar, India. The main characteristics of the APW are summarized in the Table 1. The EC process performance was determine by optimize the parameters from varying the ranges: pH (4.5–10.5), current density (j: 34.72–121.52 A/m$^2$), electrolysis time (t: 30–150 min), and salt concentration (m: 0–2 g/l). After treatment, samples were collected, filtered, and analyzed for COD, color, turbidity, final pH change, total suspended solid (TSS), metal and nonmetal ions concentration and chlorine concentration. The electrode plates were manually cleaned with 15% HCl solution before and after treatment for their reuse. Energy consumption in kWh was calculated for the removal of 1.0 kg of COD. In electrolysis, average cell voltage was used for calculating the energy consumption [22,23].

The removal efficiencies (RE) of color, COD, and turbidity were calculated by using the following relation in Eq. (1)

$$\text{Removal efficiency (RE) in percentage} = \frac{C_i - C_f}{C_i} \times 100 \quad (1)$$

Where, $C_i$ and $C_f$ are initial and final concentration of the samples at any time. The color removal efficiency was evaluated by determine the difference of solution absorbance before and after the treatment. To determine the amount of chlorine concentration with Al and SS electrodes at their optimum treatment conditions of *t*, *j*, pH and NaCl for each electrodes; the iodometric titration method was performed, where 5 ml of aliquots were sampled and analyzed chemically, as shown by Singh et al. [24] and Neodo et al. [25].

### 2.2. Analytical methods

Different equipment/instruments were used for the analysis of different parameters. Conductivity and solution pH were determine by using the multiparameter digital meter (HACH, USA). Colorimeter (Aqualytic, Germany) and turbidity meter (Aqualytic, Germany) were used for measured the color and turbidity of the solution, respectively. Double beam UV–visible spectrophotometer (HACH, DR 5000, USA) was used for determine the COD of the samples. The samples were digested in the DRB 200, HACH, USA digestion unit. Field emission scanning electron microscopy (FESEM) coupled with energy dispersive X-ray (EDX) (SEM, QUANTA, Model 200 FEG, USA) was used for determine the solid residue (sludge and scum) morphology and elements distribution in the residues. Thermo-gravimetric analysis (TGA) of solid residue was carried out from the temperature range 20 to 1000 °C in air atmosphere at 10 °C/min heating rate in a Perkin Elemer (Pyris Diamond) thermogravimetric analyzer. The metal and nonmetal ions concentration were determine by using the UV/vis detector equipped ion chromatography (IC) (Mehtrom). Before IC analysis, samples were first digested in nitric acid solution than filtered with 0.22 µm membrane filter.