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Research Article

The Efficiency of Electrocoagulation and Electroflotation Processes for Removal of Polyvinyl Acetate From Synthetic Effluent

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Abstract

Polyvinyl acetate (PVA_c) is a type of thermoplastic resin generated by a polymerization of vinyl acetate. Effluent of this polymer is highly rich with chemical oxygen demand (COD) and total solids (TS). Due to lack of studies on the above problem, the current study aimed at obtaining a sufficient method for the effluent pre-treatment. In fact, the study discussed PVA_C effluent treatment by electrocoagulation (EC) and electroflotation processes.The study considered the effect of various operating parameters such as pH and current density, initial concentration of pollutant, inter-electrode distance, electrolysis times, and types of electrode materials (iron and aluminum); COD and TS removal efficiency and optimal values of operational parameters were calculated. In the study, COD and TS reduction rates in the optimized conditions in batch flow reactor were 83% and 78%, and 80% and 72% for Fe and Al electrodes, respectively. Optimized conditions were taken as 24 mA cm $^{-2}$, 20 g/L PVAC, and neutral pH in 20 minutes for Al-Al electrodes and 15 minutes for Fe-Fe electrodes, 1 cm distance between electrodes with parallel-type monopole of connection modes. According to the results, electrochemical process with batch flow tends to be a suitable pre-treatment process that is inexpensive, easily operated, and highly sufficient for effluent treatment, which contains polyvinyl acetate.

Keywords: Effluent Treatment, Polyvinyl Acetate, Electrocoagulation, Electroflotation

1. Introduction

Nowadays, more non-biodegradable compounds enter the environment. Increased production of such combinations created concern regarding the treatment of this wastewater before discharging it into the environment (1). In industry, protection of the environment and waste recycling are very important; due to the high density of biochemical oxygen demand (BOD) and chemical oxygen demand (COD) and organic loading, these factors have the most important role in environmental pollution and generally have adverse effects on physical, chemical, and biological characteristics of the environment. Treatment of the industrial wastewater to achieve the desired environmental standards is essential. Effluent of the industries producing resin and polyvinyl acetate resin, emit high COD into wastewater refinery system of industrial parks (2, 3). Therefore, this effluent interferes with the treatment processes. Thus, a pre-treatment process is required before carrying this chemical waste to the drain of the sewerage system. Polyvinyl acetate is used as an adhesive in the textile industry and in manufacturing carpets it is used for sizing (1, 4). Polyvinyl acetate is a type of thermoplastic polymer crested during vinyl acetate emulsion polymerization by a suitable initiator without any solvent or in the presence of water or 2-propanol. PVA_C is a white powder insoluble in water (2). For the first time, vinyl acetate was discovered by a German scientist named Fritz Klatte in 1912 and polyvinyl acetate emulsion is the 1st synthetic polymer resin built on a world scale (3, 5, 6). There are a variety of methods to refine chemical effluents; some of the most widely used methods include biological treatment, chemical precipitation, and adsorption on carbon, ultrafiltration, oxidation, and ozonation (7). The urgent need for more effective and less expensive methods for wastewater treatment is well known. Electrochemical treatment process is a promising and developing technology (8, 9).

The main removal mechanisms in electrochemical process include electrocoagulation, electroflotation, electrooxidation, and every combination of these mechanisms (10, 11). Reactors of this process are called electrolysis cells. Each cell is formed of a multielectrode as anode and cathode, an electrolyte, and a tank; where they occur in the process (12). The difference between the electrocoagulation and chemical coagulation is mainly the releasing of ions of coagulation factor of the method. In the electrocoagulation flocculation and sedimentation do not take place through the release of chemical coagulants into the system, yet it is performed by the electrodes in the reactor (13). The removal mechanism of PVAc is through electrocoagulation, coagulation, and flotation using iron and aluminum electrodes. In this process, clots start floating and, then, are settled and finally become separated from the effluent (14). These processes can be used effectively due to the high performance, easy manufacturing, low manufacturing costs, and low energy consumption compared to the traditional methods (15). EC process involves the generation of positive ions (Al^{3+} and Fe^{2+}) in the anodes, in the solution of wastewater, and production of hydrogen gas in the cathode at the same time:

In anode:

$$M \rightarrow M^{n+}_{(aq)} + ne^{-} \tag{1}$$

$$2H_2O \rightarrow 4H^{+}_{(aq)} + O_{2(g)} + 4e^{-} \tag{2}$$

In cathode:

$$M^{n+}_{(aq)} + ne^{-} \rightarrow M_{(s)} \tag{3}$$

$$2H_2O_{(I)} + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-}_{(aq)} \tag{4}$$

In this mechanism M represents the intended electrode and n is the number of electrons. In electrocoagulation, coagulation of ions is produced at site and generation process includes these consecutive steps as dissolution of the anode- formation of OH ions, H_2 at the cathode-electrolytic reactions on the surfaces of electrodes- formation of polymers and monomers (responsible for destabilizing colloids), and removal of keloids by sedimentation and flotation (16, 17).

The increasing number of polyvinyl acetate manufacturers and growing trend of this industry in the country on one hand, and the lack of enough studies regarding the effluent treatment of these industries on the other hand prompted authors to study the efficiency of polyvinyl acetate effluent treatment through electrochemical process. The study considers the effect of operating various parameters such as: pH and current density, initial concentration of pollutant, inter-electrode distance, electrolysis times, and types of electrode materials (iron and aluminum); COD and total solids (TS) removal efficiency and optimal values of operational parameters.

2. Materials and Methods

It was a laboratory experimental study conducted in batch flow reactor in water and wastewater chemistry laboratory, Faculty of Health, Hamadan University of Medical Sciences, Iran. This laboratory study was conducted in batch mode to investigate the efficiency of electrocoagulation and electro flotation processes to remove polyvinyl acetate in synthetic effluent. The characteristics of PVA_C are listed in Figure 1. Electrocoagulation and electroflotation processes were conducted in a 400-mL reactor. A pair of anodes and a pair of cathodes with dimensions of 8 × 2.5 cm and electrode connection of parallel-monopole were used and connected to a direct current (DC) power supply (Model Dazheng 305D, China). Height of 5 cm of electrodes dipped in synthetic wastewater containing polyvinyl acetate. During the tests, the reactor was placed on a magnetic stirrer with the speed of 100 rpm for mixing operation. Table 1 and Figure 2 show the reactor, and the properties and characteristics of the synthetic effluent. Before starting each experiment, the electrodes were abraded with sand paper to remove scale, in 3N-hydrochloric acid for 10 minutes (18). All experiments were performed at room temperature. To increase the concentration of ions and decrease the electrical resistance in decomposition solution, 0.5 mg of NaCl was added to the solution as supporting electrolyte; also, 0.5 N sulfuric acid and 0.5 N sodium hydroxide were used to adjust the pH. The pH values were measured using a pH meter (Hach SensION1, Germany). Since the study mainly aimed to apply tests under a constant current density condition, the current density through reaction was fixed and voltage changes were recorded. Flocs and coagulants required a period of time for coagulation and flocculation; thus, 60 minutes were allowed at the end of each experiment, then, samples were taken. COD and total suspended solids (TSS) experiments were measured according to standard methods (19). COD was measured with the "5220C" Close Reflux .Samples for COD measurement were put into the reagent-containing vials ($HgSO_4$, $H2Cr_2O_7$, Ag_2SO_4, and H_2SO_4) and incubated at 150°C for 2 hours. Then, the absorbance of reagent was determined by DR-5000 Spectrophotometer (Hach, Germany); according to the standard curve of COD to determine the COD concentration. TS was measured by gravimetric analysis. The TS and COD removal efficiencies were calculated as:

$$(R\%) = \left(\frac{C_i - C_t}{C_i}\right) \times 100 \tag{5}$$

Where, C_i and C_t are initial and final concentrations of COD and TS (as mg L^{-1}) (20).

3. Results and Discussion

Effluent treatment by electrocoagulation and electroflotation is a complex process that depends on operating parameters such as: pH and current density, initial concentration of pollutant, inter-electrode distance, electroly-

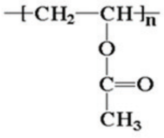
Molecular Formula	$[CH_2CHCOOCH_3]n$
Molecular Weight (Da)	60000-100000
Molecular Structure	
Density (g/mL)	1.10
Boiling Point (°C)	112
Percent Solids (at 105°C)	1 ±50
Viscosity (m pa/s) (at 22 ° C)	85000 ± 15000

Figure 1. Specifications of Polyvinyl Acetate

Table 1. Specifications of Synthetic Containing Polyvinyl Acetate (20 mg/L)

Parameters	Value
COD, mg/L	18 000
pH	6.5
Electrical Conductivity, μS/cm	349
TDS, mg/L	176
Temperature, °C	21.2

sis times, and types of electrode materials (21). In the current study, 0.5 mg of NaCl was added to the solution to supply the electrical conductivity. Studies show that this supporting electrolyte has negligible effects on the initial pH (about 0.3), when using an iron electrode and an aluminum electrode (22).

3.1. Effect of pH

Studies show that pH is an important influencing factor in electrochemical processes. The initial pH determines the type of production of metal ions and affects the type of ions in solution and solubility products (23). Synthetic effluent with the initial pH values in the range of 3 to 11 was used to investigate the effect of initial pH and pH variations during the synthetic effluent on COD and TS removal efficiency by aluminum and iron electrodes. Figure 3 shows that removal efficiencies of COD and TSS are the function of pH. The highest efficiency was calculated in both electrodes at neutral pH (6 - 7), the pH of synthetic effluent. Therefore, this process was not a need to optimize pH. Figure 3 also presents final pH. Final pH changes after electrocoagulation as a function of initial pH and electrodes material (23). When using iron electrodes, the final pH increases and is always higher than the initial pH. But when using aluminum electrodes, in initial pH greater than 7, the final pH decreases and at a pH of 7 and lower, pH increases and it is similar to the results reported by the Kabdaslı I, et al. This situation shows that pH changes during the electrochemical process. Due to the production of hydroxide ions during the process of reduction of water molecules at the cathode electrode, pH usually increases. In alkaline conditions, $Al\ (OH^-)_4$ complex is the key to reduce pH. Hydrogen bubbles produced at the cathode at neutral pH are small and thin, but they can be used to create an area to connect liquid, gas, and solid, and accumulate small neutral and colloidal particles. However, different balances occur at aluminum electrodes between hydroxide that depend on the PH and buffering properties of the waste (24, 25).

3.2. Effect of Current Density

According to the mathematical equation, current density is a result of dividing electric current by effective area of anode electrodes. According to previous studies in the batch reactor of electrocoagulation, current density is a vital operating factor; in fact, current density is an essential parameter that controls the process. It is well known that DC affects production of coagulant dose rate, position and rate of cation dosing, bubble production and bubble path length, and controls reaction speed in the reactor (9). In the current study, optimization of current density was studied in constant charge passed (14.4 C/cm^{-2}) at different current densities (6, 12, 18, 24, and 30 $mA\ cm^{-2}$) and the effect of current density was determined by measuring COD and TS removal efficiency. According to the Faraday law in constant effective area, the relationship between reaction time and current density is reversed (26). Therefore, by increasing current density, the required time for the EC process is reduced. When the current density is 6 $mA\ cm^{-2}$, electrolysis time is 40 minutes and with the increase of current density to 30 $mA\ cm^{-2}$, electrolysis time is 8 minutes. When the current density is enough in the solution, produced metal ions are hydrolyzed by dissolution of the sacrificial anode and converted into a series of metal hydroxide. The metal hydroxides neutralize hydrostatic loads on dispersed particles. As a result, electrostatic forces of repulsion between charged particles are effectively reduced and

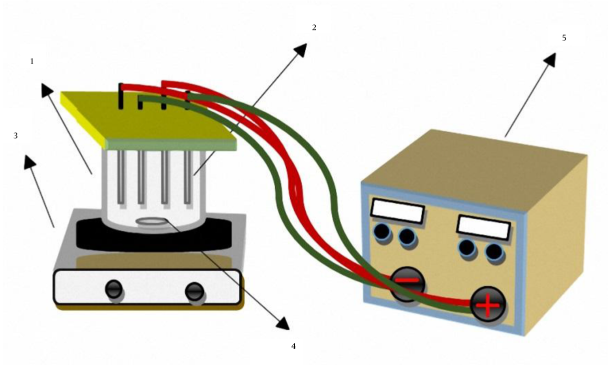


Figure 2. Experimental setup used in the EC process (1. Batch EC reactor 2.electrodes 3.magnet stirrer 4.magnet 5. Power supply)

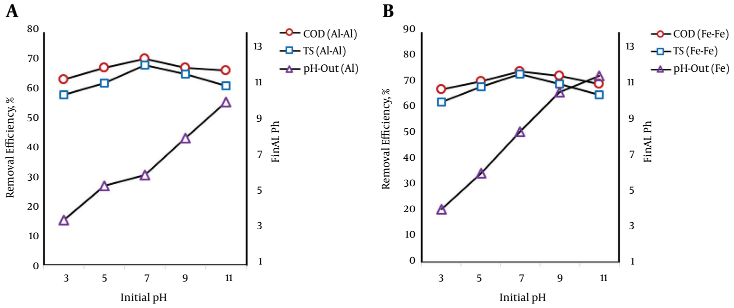


Figure 3. Effect of pH on COD and TS Removal Efficiency (%) by Al-Al (A) and Fe-Fe Electrodes (B) and pH Variation During Electrocoagulation and Electroflotation Processes (Electrolysis Time = 20 Minutes, Current Density = 12 mA cm^{-2}, and Concentration (PVA_C) = 20 g/L)

the Van der Waals attractive forces prevail and particles of aggregation and flocculation easily become possible (26). In the study, in higher current density and less electrolysis time, process efficiency was better. But as the EC process needs a minimum period, in the less than its, the efficiency is very low (27, 28). In the study, when the current density was 30mA cm^{-2} the efficiency reduced since the electrolysis time reduced to less than 10 minutes. As Figure 4 shows, in both electrodes, the highest percentage removal efficiency COD and TS were obtained. In a current density of 24 mA cm^{-2}, iron electrodes had greater efficiency, in comparison.

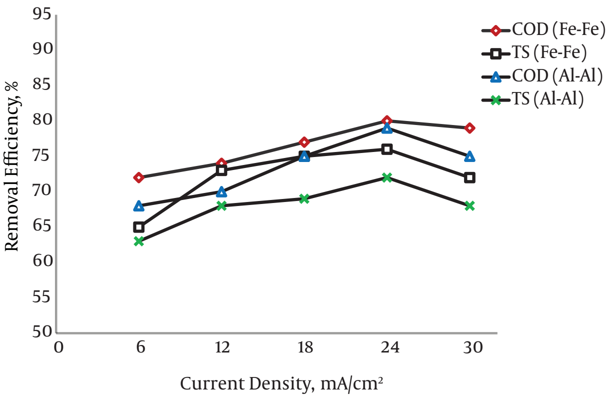


Figure 4. Effect of Current Density on COD Removal Efficiency (%) by Al-Al and Fe-Fe in Electrocoagulation and Electroflotation Processes (Charge Passed = 14.4 C/cm^{-2}, Concentration (PVA_C) = 20 g/L, and pH = Neutral)

3.3. Effect of Electrolysis Time

Figure 5 shows the results of TS and COD removal efficiency (%);the highest TS and COD removal efficiency (%) was achieved in 15 minutes when using iron electrodes; while, by aluminum electrodes the achieved efficiency was different and the duration was 20 minutes. Thus, the required time to achieve the highest efficiency to reduce with iron electrodes was less than that of aluminum electrodes. By applying iron and aluminum separately and respectively for 15 and 20 minutes, the COD and TS removal efficiency decreased. In a similar study, Elneny AMH et al. achieved similar results (29). According to the conducted studies, it can be interpreted that the metal hydroxides are produced by dissolution of the anode and with constant current density, increase of time increases the number of metal hydroxides. By increasing time, the flocs have more opportunity for flocculation or sedimentation. The efficiency does not increase; in spite of abstaining the required time it continues until the desired time and beyond that as dissolved ions are reduced and the production of flocs decrease (30, 31). It should be noted in the current research that although the difference between the efficiency in 2 electrode materials was negligible, the efficiency of iron electrodes was 2% more. In this process, organic molecules of polyvinyl acetate are trapped by Fe $(OH)_3$ and Al $(OH)_3$ ions, and other monomers and polymers produced from hydrolysis, and are removed from the effluent through the sedimentation and being trapped in the bubbles produced from production of H_2 in the cathode. COD and TS removal efficiency in electrocoagulation can be attributed to the removal of dangling solids and flow of liquid organic molecules as metal organic combinations. However, it is necessary to conduct meticulous studies on oxidation reactions, reduction on the surface of the electrodes, and the bulk phase formed to create the mechanisms of electrochemical removal of COD (32).

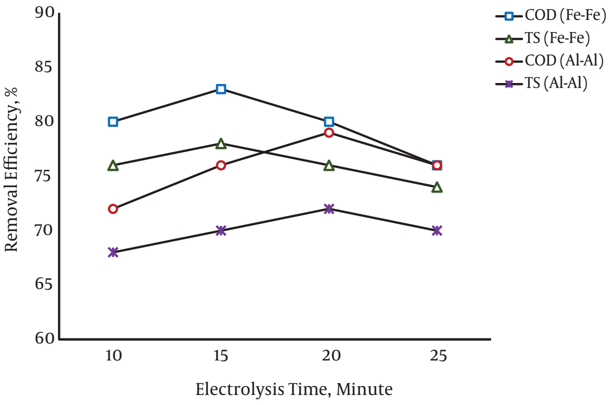


Figure 5. Effect of Time on COD and TS Removal Efficiency (%) in Electrocoagulation and Electroflotation Processes (Current Density = 24 mA cm^{-2}, Concentration (PVA_C) = 20 g/L, and pH = Natural)

3.4. Optimization of Inter-Electrode Distance

Many studies indicated the effect of the inter-electrode distance in electrochemical processes. Based on these studies, the distance between the electrodes is an important parameter in electrocoagulation. And the evolution of performance of electrochemical processes is a common function of the combination of distance between the electrodes, pollutant nature of the electrode structure, and dynamic characteristics and other factors, and also the potential Ohmic-reduction in the electrochemical cell is proportional to the inter-electrode distance. Inter-electrode distance affects the energy consumption of electrolysis, especially when samples have low electrical conductivity (11, 33). In the current study, to optimize the condition of other parameters, the distance between electrodes was studied at 1-, 2-, and 3-cm levels (Figure 6). As the shows, shows, the best results were obtained at a distance of 1-cm which was in line with those of Najer Samir Ahmed et al. Increasing the distance reduces the attraction between the electrodes and the present ions in the dissolved solution. Hence the flocculation process is done slowly (34).

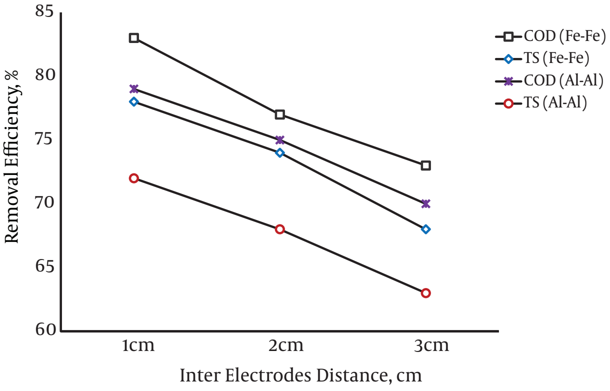


Figure 6. Effect of Inter Electrodes Distance on COD and TS Removal Efficiency (%) Under Optimal Conditions of Electrocoagulation and Electroflotation Processes (Electrolysis Time = 20 Minutes for Al-Al Electrodes and 15 Minutes for Fe-Fe electrodes and, Current Density = 24 mA cm^{-2}, Concentration (PVA_C) = 20 g/L and pH = Natural)

3.5. Optimization of Concentration of the Pollutant

In the current study, densities of 5, 10, 15, 20, 25, 30 g L^{-1} polyvinyl acetate were examined. Figure 7 shows the effect of different densities on aqueous solutions. The findings showed that by increasing concentration of PVA_C in aqueous solution efficiency, COD and, TS reduced and flocculation and the size of flocs increased. But, as density of 5 g L^{-1} PVA_C got closer to 20 g L^{-1}, the increase was not noticeable. According to the Faraday law, in the constant current density and electrolysis time, in every density of pollutant, the amount of dissolved metal hydroxide entered via the anodes is fixed. For this reason, the amount of metal produced from hydroxyl ions on the surface of electrodes, in high concentrations of PVA_C, is not sufficient. Therefore, as stated Al Aji B et al., to obtain higher efficiency in high densities, the current density and electrolysis time should also be increased (35).

4. Conclusion

The study found that electrocoagulation and electroflotation for pre-treatment of effluent containing

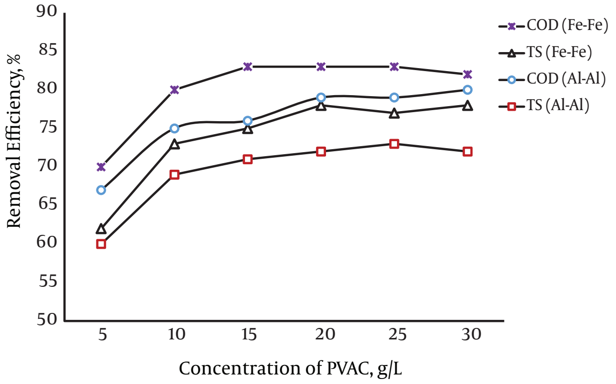


Figure 7. Effect of Concentration of PVA$_C$ on COD Removal Efficiency (%) in Electro-Coagulation and Electro-Flotation Processes (Electrolysis Time = 20 Minutes for Al-Al Electrodes and 15 Minutes for Fe-Fe Electrodes, Current Density = 24 mA cm^{-2}, and pH = Natural)

polyvinyl acetate is efficient and COD and TS removal efficiency increase by increasing the current density and electrolysis time. However, it is necessary to determine the optimum point to reduce operating costs and increase refinement efficiency to achieve environmental standards. In the process, according to the reduction percentage of COD and TS, iron electrodes efficiency was higher than that of aluminum. It should be noted that this efficiency difference was negligible. Therefore, the implementation of the process with respect to the further corrosion of aluminum compared to metal electrodes, this difference can be ignored. The results showed that electrocoagulation and electroflotation processes in initial pH of the effluent by iron electrodes in concentration of 20 g L^{-1} of PVA$_C$ during 15 minutes, current density 24 mA cm^{-2} and distance of 1 cm between the electrodes was reduced to 83% of COD and 78 % of TS. According to the results, electrochemical process with batch flow tends to be a suitable pre-treatment process that is inexpensive, easily operated, and highly sufficient for effluent refinement that contains polyvinyl acetate.

Acknowledgments

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