Electrocoagulation (Ec) of Styrene Butadiene Rubber from Aqueous Solution using Different Electrodes
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Abstract:
The removal of Styrene Butadiene Rubber (SBR) from aqueous solution by electrocoagulation using different electrodes was investigated. The main objectives of the experiments were to investigate the effects of the various operating parameters on the SBR and chemical oxygen demand (COD) removal efficiency from aqueous solution. The optimal operating condition for removal of (SBR) were: initial pH of 7.1, an initial polymer concentration of 200 mg/L, temperature of 30 oC, current density (25 mA/cm2) and salt concentration of (1.5 g/L) by using iron (Fe), stainless steel (SS) and aluminum (Al) electrodes. The results showed that the removal percentage for SBR and COD were (99.2% and 91.2 %), (99.4% and 93.5%) and (98.9 % and 90.2%) by using (Fe), (SS) and (Al) electrodes at 15 min respectively. The adsorption equilibriums were analyzed by Langmuir and Freundlich isotherm models. It was found that the data fitted to Freundlich (R² = 0.954) better than Langmuir (R² = 0.777) model. The removal of SBR exhibited a pseudo first order reaction with rate constant (0.191, 0.245 and 0.142 min-1) for Fe, SS and Al electrodes respectively.

Keywords : Styrene Butadiene Rubber, Electrocoagulation, Water Treatment, Electrode, Adsorption, Kinetics

Introduction:
Electrocoagulation (EC) is an emerging technology that combines the functions and advantages of conventional coagulation, flotation, and electrochemistry in water and wastewater treatment. Each of these fundamental technologies has been widely studied separately. However, a quantitative appreciation of the mechanism of interaction between these technologies employed in an electrocoagulation system is absent [1]. A literature survey indicates that EC is an efficient treatment process for different wastes, e.g. soluble oils[2], liquid from the food[3], textile industries [4] and effluents from the paper industry [5]. EC is an effective process for the destabilization of finely dispersed particles by removing hydrocarbons, greases, suspended solids and heavy metals from different types of wastewater [5]. The possible reactions which occur during the electrocoagulation process are [6 -8]:

At the anode:
M → M n+ + ne- (1)

At the cathode:
2 H2O → 4H+ (aq) + O2 (g) + 4e- (2)
Mn+(aq) + ne - → M (s) (3)
H2O(l) + 2e- → H2 (g) + 2 OH- (aq) (4)

Where M is the material used as electrode and n is the number of electrons. During the electrocoagulation process metal hydroxides, polyhydroxides and/or polyhydroxymetallic compounds of the electrode material will be generated. These materials contain strong affinity for dispersed particles and counter ions, which results in coagulation [6].

Styrene butadiene rubber (SBR) is one of the earliest industrial synthetic rubbers and has the largest consumption among various general-purpose synthetic rubbers. SBR is the random copolymer of styrene and butadiene, the overall performance of which is similar to that of natural rubber (NR). Some properties of SBR...
such as abrasive resistance, heat resistance, and aging resistance are superior to those of NR, and thus SBR has replaced NR partially in many fields. However, due to the instability of double bonds in SBR to ultraviolet, heat, and oxygen, aging processes like chain scission, chain crossing, and oxidization take place easily when the SBR products are in use just like other styrene–butadiene copolymers [9–16].

Various treatment procedures have been reported for the removal of Styrene butadiene rubber from water e.g. Thermal oxidative [17], Photooxidation and [18], microwave energy [19], catalytic oxidation [20], coagulation-catalytic oxidation Process [21] and Fenton reagent [22].

The purpose of the present work is to study the performance of EC process on the removal of Styrene butadiene rubber and COD from aqueous solution using iron (Fe), stainless steel (SS) and aluminum (Al) electrodes and to investigate the kinetic and adsorption isotherm studies on the removal efficiency.

Experiment:

Chemicals:
The polymer used in the present work was Styrene butadiene rubber, polymer solutions were prepared from the commercially available polymer in distilled water at a concentration of 500 mg L–1. The properties of the Styrene butadiene rubber is given in Table 1. Sodium chloride, sodium sulfate, sodium carbonate, potassium chloride, sodium fluoride, sodium hydroxide, sulfuric acid, potassium dichromate, were of analytical grade and purchased from Merck. Distilled water was used for the preparation of solutions. Standard solutions of potassium dichromate (K2Cr2O7), sulfuric acid (H2SO4) reagent with silver sulfate (Ag2SO4), Mercury sulfate (HgSO4) and sulfuric acid were prepared to measure the COD. A stock solution of polymer (500 mg/L) was prepared by dissolving an accurate quantity of the polymer in distilled water and suitably diluted to the required initial concentrations. Different standard solutions of polymer with concentration from 50-200 mg L–1 were prepared to measure its removal at different conditions. The pH of the working solution was adjusted to the desired values with 0.1N HCl or 0.1N NaOH.

Properties of SBR

| Polymer | Styrene butadiene rubber or Buna S |
|---------|-----------------------------------|
| IUPAC Name | buta-1,3-diene:styrrene |

Chemical structure

![Chemical Structure of SBR](image)

| Chemical formula | (C12H14)n |
|-----------------|----------|
| Water solubility | 8.1–8.6 g/L |
| Physical Appearance | Milky with Bluish Emulsion |
| Viscosity | 100–350 mPa |
| Molecular weight (g/mol) | 69,700 g/mol |
| λ_max | 305 nm |

**Table 1:** Properties of Styrene butadiene rubber
Equipments and procedures:
The electrocoagulation unit consisted of an 100 ml electrochemical reactor used SS with SS , Fe with Fe and A1 with A1 electrodes as (anode and cathode) with an effective surface area of 4 cm². The electrodes were 20 mm×10 mm and inter electrodes distance was 1 cm. The electrodes were positioned vertically and parallel to each other . The current density was maintained constant by means of a precision DC power supply, model (DZ040019) EZ DigitalCO. Ltd. (Korea). The polymer concentration was determined using a double-beam UV-Vis spectrophotometer, model UV 1601 is from Shimadzu (Japan) at 305 nm. Hot Plate , model (HB502), BIBBY STERILIN LTD .(U.K.) . A pH meter model AC28, TOA electronics Ltd., (Japan) . Water bath model SB-650 , Tokyo Kikakkai CO. Ltd. , (Japan). A closed reflux titrimetric unit was used for the COD determination. Chemical Oxygen Demand (COD), HANNA instruments ,Thermo reactor , model C9800 Reactor in Hungary – Europe.

Analysis:
Two main parameters were measured to evaluate the electrochemical treatment efficiency, the remaining pollutant concentration and the COD. Remaining pollutants styrene butadiene rubber concentration was measured with the double-beam UV-visible spectrophotometer at λmax= 305 nm using calibration curve with standard error ±0.5%. The COD was determined using a closed reflux colorimetric method. The equation used to calculate the polymer removal efficiency in the treatment experiments was:

\[ \%E = \left[ \frac{(A_0 - A)}{A_0} \right] \times 100 \]  

(5)

Where A₀ and A are absorbance values of polymer solutions before and after treatment with respect to their λmax [23].

The calculation of COD removal efficiencies after electrocoagulation treatment were performed using the following formula [24].

\[ CR(\%) = \left[ \frac{(C_0 - C)}{C_0} \right] \times 100 \]  

(6)

Where C₀ and C are concentrations of polymer before and after electrocoagulation.

Result and Discussion:
Effect of current density (mA/cm²):
The current density (CD) is the amount of current per area of the electrode. It is an important parameter in removal efficiencies [25]. In all electrochemical processes, current density is the most important parameter for controlling the reaction rate within the electrochemical reactor [23]. To examine the effect of current density on SBR and COD removal efficiency, a series of experiments were carried out with the current density ranging from (10 - 50 mA/cm²) at a pH of 7.1, initial concentration of 200 mg/L, , NaCl concentration of 1.5 g/L and temperature of 30 oC. From Figure. 1 and table 2 indicated that the optimum current densities were 25 mA/cm² for Fe, S-S and Al electrodes. The removal efficiencies of SBR were 99.2%, 99.4% and 98.9% while the COD removal efficiencies were 91.5%, 93.5% and 90.2% using Fe, S.S and Al electrodes at 15 min respectively. In general the current density decreased, the time needed to achieve similar efficiencies increased. If current density increased the % removal of contaminants also increased. The increasing of current density always increase the cost of treatment, so it is necessary to select an optimum value of current density for efficient treatment and minimum cost.

Fig.1: Effect of current density on the efficiency of Styrene butadiene rubber adsorption using iron (a), stainless steel (b) and aluminum (c) electrodes respectively
Table 2: Effect of current density, pH, type of electrolyte, concentration electrolyte, rubber concentration, and temperature on the efficiency of COD removal for Styrene butadiene rubber using iron (a), stainless steel (b) and aluminum (c) electrodes respectively.
Effect of initial pH:
The pH is one of the important factors affecting the performance of electrochemical process. The pH value increased as the operating time of EC process was increased due to the OH- ion accumulation in aqueous solution during the process [26]. To examine the effect of initial pH on removal efficiency, a series of experiments were performed on 200 mg/L of SBR solution with different initial pH in the range of (3-11). Figure 2 and table 2 show that the highest removal efficiency of the SBR and COD can be achieved near neutral (PH7.1) using all working electrodes. The lower removal efficiencies at higher acidic and alkaline pH can be explained by amphoteric behavior of metal hydroxides which lead to soluble cations and to monomeric anions [27].

![Fig. 2: Effect of pH on the efficiency of Styrene butadiene rubber adsorption using iron (a), stainless steel (b) and aluminum (c) electrodes respectively.](image)

Effect of electrolyte concentration (g/L):
Electrolyte is a substance which is responsible for increasing the conductivity of the solution. The effect of NaCl concentration on removal efficiencies of SBR and COD were investigated at time 15 min, initial concentration of 200 mg/L, pH of 7.1, temperature of 30 oC and current densities of 25 mA/cm2 using Fe, S-S and Al electrodes. Figure. (3) and table 2 show that the maximum removal efficiencies of SBR and COD was obtained at 1.5 g/L NaCl concentration. Increase in the concentration of the electrolyte results an increase in number of ions. So, ultimately solution becomes more conductive and the passed current increases and the produced amount of metallic hydroxide and SBR removal increases [28]. However, with the increase in NaCl concentration >1.5 g/L the removal efficiency decreased.
Effect of initial SBR concentration (mg/L):
The effect of initial SBR concentration on the removal was examined with solutions including polymer of 50, 100, 150 and 200 mg/L at time 15 min, pH of 7.1, temperature of 30°C and current densities of 25 mA/cm² using Fe, S-S and Al electrodes. According to the Figure 4 and Table 2, it may be seen that increasing initial polymer concentration results in increasing removal efficiency. One of the most important pathways of SBR removal by electrocoagulation is adsorption of SBR molecules on metallic hydroxide flocs. The adsorption capacity of flocs is limited and specific amount of flocs is able to adsorb specific amount of SBR molecules [29].
Effect of temperature (°C):
The effect of varying temperature from 10 to 40°C has been studied for the SBR removal efficiency and COD at initial SBR concentration of 200 mg/L, time of 15 min, pH of 7.1 and current densities of 25 mA/cm² using Fe, S-S and Al electrodes. Figure 5 and Table 2 indicate that the removal efficiency of SBR and COD increase with the increasing temperature up to 30°C due to increasing diffusion and the motilities of ions. Further increasing of temperature higher than 30°C has a negative effect on removal efficiency of SBR and COD. The polymer and COD removal efficiency dropped to low values. This attributed to the volume of the colloid M(OH) will decrease and pore production on the metal anode well be closed [30].
Fig. 5: Effect of temperature on the efficiency of Styrene butadiene rubber adsorption using iron (a), stainless steel (b) and aluminum (c) electrodes respectively.

Effect type of electrolyte:
Figure 6 and Table 2 explain the effect of electrolyte type on the removal efficiency of SBR and COD using Fe, SS and Al electrodes. In the presence of different supporting electrolytes including NaCl, KCl, Na2SO4, NaF and Na2CO3, was studied at initial concentration of 200 mg L-1, a current density of 25 mA /cm2, inter electrode distance of 1 cm, a temperature of 30°C and pH of 7.1. According to Figure 6 and Table 2 the SBR and COD removal using NaCl electrolyte is better than other electrolytes (KCl, Na2CO3, NaF and Na2SO4). Due to formation of hypochlorite (OCl-) and hypochlorous acid (HOCl). It is well known that Cl– anions can destroy the formed passivation layer on electrode and therefore enhance anodic dissolution rate of metal which lead to produce more metal hydroxide [31,32].
Application of paint in SBR polymer treatment from aqueous solution:
The treatment of SBR co-polymers effluents obtained from Sizer spuc paint was carried out by using Fe, SS and Al electrodes. The initial polymer concentration of these sample was (10-12%) for SBR taken from Sizer co. for paint production paints located in Gaza Strip- Gaza City. 200 mg/L of paint solutions were treated by the electrocoagulation technique using the same electrodes under the optimum condition. After the treatment process, the removal percentages of SBR polymer from its paint at 15 min using Fe, SS and Al electrodes were (72.1%, 67.9%, and 62.1%) respectively. The removal percentages of SBR COD were (68.1%, 64%, and 60.2%) respectively. These results indicated that the suggested studied electrodes are highly efficient in the treatment of effluents containing SBR co-polymer with very slight effect of matrix [33]

Kinetics studies:
The kinetic study is an important issue in order to examine the order of the reaction[26]. In an attempt to throw some light on the kinetics of the present process which is difficult to quantify owing to its complexity and it is being controlled by many interacting variables. Figure 7 represents the removal of SBR exhibited pseudo first order with good correlation coefficient 0.979, 0.992 and 0.946 using Fe, S-S and Al electrodes according to following equation:

\[
\ln A_t/A_0 = -kt
\]

Where \( A_0 \), \( A_t \), \( t \), and \( k \) are the polymer absorbance at initial concentration, polymer absorbance at each time, time of reaction (min), and reaction rate constant, respectively. The calculated \( k \) values from Figure. 7 are 0.191, 0.245 and 0.142 min\(^{-1}\) using Fe, S-S and Al electrodes respectively. Results show that the removal rate using SS electrode was higher than the removal rate using Fe and Al electrodes.

Fig. 6: Effect of type of electrolyte on the efficiency of Styrene butadiene rubber adsorption using iron (a), stainless steel (b) and aluminum (c) electrodes respectively.
Fig. 7: Relation between Ln At against the time for Styrene butadiene rubber adsorption using iron (a), stainless steel (b) and aluminum (c) electrodes respectively.

**Adsorption isotherm:**

**Freundlich Adsorption Isotherm:**
It is empirical model relating the adsorption intensity of the sorbent towards adsorbent. The isotherm describes the multilayer adsorption with a heterogeneous energetic distribution of active sites, accomplished by interaction between adsorbed molecules [34]. The linearized and logarithmic expression of the Freundlich model is [35].

\[
\log q_e = \log k_f + n \log C_e \tag{8}
\]

where \( k_f \) (mg/g) and \( n \) (dimensionless) are constants that account for all factors affecting the adsorption process, such as the adsorption capacity and intensity. The Freundlich constants \( k_f \) and \( n \) are determined from the intercept and slope, respectively, of the linear plot of \( \log q_e \) versus \( \log C_e \).

**Langmuir Adsorption Isotherm:**
The Langmuir isotherm assumes monolayer deposition of adsorbate on homogenous adsorbent surface. It is well known that the Langmuir equation is intended for a homogeneoussurface [34]. The linearized form of Langmuir adsorption isotherm model is [36].

\[
C_e/q_e = 1/q_m b + C_e/q_m \tag{9}
\]

where \( q_e \) (mg/g) is amount adsorbed at equilibrium, \( C_e \) (mg/L) equilibrium concentration, \( q_m \) is the Langmuir constant representing maximum monolayer adsorption capacity, and \( b \) is the Langmuir constant related to the energy of adsorption obtained in terms of equations (8) and (9) by using experimental adsorption results in the above equations. The values \( q_m \), \( b \), \( K_F, RL \) and \( n \) are summarized in Table 3.
Table 2: Parameters of Langmuir and Freundlich isotherm constants and correlation coefficients using SS electrodes.

| Isotherm       | $R^2$ | Parameter          | Value  |
|----------------|-------|--------------------|--------|
| Langmuir       | 0.777 | $q_m$ (mg/g)       | 25     |
|                |       | $b$ (L/mg)         | 0.0265 |
|                |       | $R_L$              | 0.158  |
| Freundlich     | 0.954 | $K_F$ (mg/g)       | $3.47 \times 10^{11}$ |
|                |       | $n$                | 0.181  |
|                |       | $1/n$              | 5.524  |

**Energy consumption:**

In an electrochemical process, the most important economical parameter is energy consumption $E$ (kWh/m3) [37, 38]. This parameter is calculated from the following expression:

$$E = \frac{I V t \cdot 1000}{\text{volume}} \quad (10)$$

where $V$, $I$ and $t$ stand for average voltage of the EC system (V), electrical current intensity (A) and reaction time (h), respectively.

![Langmuir plot](image)

**Fig. 8:** Langmuir plot ($Ce/qe$ vs. $(Ce)$ for Styrene butadiene rubber adsorption using S.S electrodes.

![Freundlich plot](image)

**Fig. 9:** Freundlich plot (log $qe$ vs. log $Ce$) for Styrene butadiene rubber adsorption using S.S electrodes.
Table 4.: Comparison between the Electrocoagulation method for removal of SBR with other methods.

| Type of degradation                          | Reference                        | Removal % | Time |
|---------------------------------------------|----------------------------------|-----------|------|
| fenton reagent method                       | Zhang et al. 2012 [22].          | 55%       | 30 min |
| microwave irradiation                      | Seghar et al. 2015[19]           | 50%       | 24 h  |
| catalytic oxidation                         | Lunhua, 2005 [21]               | 37.7%     | -     |
| coagulation-catalytic oxidation process     | (Qing et al., 2006) [21]         | 83.1%     | -     |
| Electrocoagulation                          | present study using S.S electrodes | 93.5%     | 15 min |
| Electrocoagulation                          | present study using Fe electrodes | 91.2%     | 15 min |
| Electrocoagulation                          | present study using Al electrodes | 90.2%     | 15 min |

Table 4.: Comparison between the Electrocoagulation method for removal of Styrene butadiene rubber with other methods.

**Conclusion:**

The removal efficiency of Styrene butadiene rubber from aqueous solution was examined by electrocoagulation using iron (Fe), stainless steel (S.S) and aluminum (Al) electrodes. The effects of initial pH, initial Styrene butadiene rubber concentration, current density, type electrolyte, salt concentration, and temperature were investigated on removal efficiency and COD. It was observed that these variables significantly affected the polymer removal efficiency. The optimum SBR removal was obtained with typical operating conditions: an initial pH of 7.1, an initial SBR concentration of 200 mg/L, current density 25 mA/cm², salt concentration of 1.5 g/L and temperature of 30°C, the results showed that SBR and COD removal were (99.2% and 91.2 %), (99.4% and 93.5%) and (98.9 % and 90.2%) by using (Fe), (SS) and (Al) electrodes at 15 min respectively. The removal of polymer was exhibited pseudo first order with rate constant (0.191, 0.245 and 0.142 min⁻¹) for Fe, SS and Al electrodes respectively. The equilibrium adsorption behavior is analyzed by fitting the isotherm models of Langmuir and Freundlich. It was noted that the data fitted to Freundlich (R² = 0.954) better than Langmuir (R² = 0.777) model.

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Review of pollutants

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