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Electrochemical degradation of trichloroethylene in aqueous solution by bipolar graphite electrodes

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Abstract

In this study, we tested the use of the bipolar electrodes to enhance electrochemical degradation of trichloroethylene (TCE) in an undivided, flow-through electrochemical reactor. The bipolar electrode forms when an electrically conductive material polarizes between feeder electrodes that are connected to a direct current source and, therefore, creates an additional anode/cathode pair in the system. We hypothesize that bipolar electrodes will generate additional oxidation/reduction zones to enhance TCE degradation. The graphite cathode followed by graphite anode sequence were operated without a bipolar electrode as well as with one and two bipolar graphite electrodes. The system without bipolar electrodes degraded 29% of TCE while the system with one and two bipolar electrodes degraded 38% and 66% of TCE, respectively. It was found that the removal mechanism for TCE in bipolar mode includes hydrodechlorination at the feeder cathode, and oxidation through reaction with peroxide. The results show that the bipolar electrodes presence enhance TCE removal efficiency and rate and imply that they can be used to improve electrochemical treatment of contaminated groundwater.

Keywords

trichloroethylene; groundwater; bipolar electrodes; treatment

1. Introduction

Trichloroethylene (TCE) was first detected in groundwater in 1977 and has been one of the most frequently detected priority contaminants in groundwater at hazardous waste sites in the United States [1]. The primary sources that release TCE into the environment are metal cleaning and degreasing operations. At many TCE spill sites, residual amounts of TCE persist in a pure liquid phase (commonly referred to as dense, non-aqueous-phase liquids, DNAPLs) within pore spaces or fractures. The dissolution of residual TCE results in a contaminated plume of groundwater (USEPA 2007). Because of potential carcinogenic and mutagenic effects [2], the Maximum Contaminant Levels (MCLs) for TCE in drinking water

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is set to $5 \mu\text{g L}^{-1}$. In addition, vapor intrusion into building has been a growing concern in recent years at sites with TCE contamination in soil or groundwater.

Methods to remove TCE and other chlorinated organic chemicals (COCs) from groundwater include microbial degradation [3–5], photochemical oxidation [6, 7], sonochemical processes [8, 9], chemical reduction via zero-valent iron [10–13], palladium-based materials [14–16] and a combination of UV with ozone, hydrogen peroxide, Fenton's reagent, or oxalate-complexes [17–21].

Electrochemical treatment has a significant advantage over other treatments since the electrochemical processes can be controlled in order to generate reducing and/or oxidizing conditions to transform contaminants [16, 22–31]. Reduction via hydrodechlorination (HDC) is favorable degradation path for TCE from groundwater [23, 25, 26, 32–34]. HDC occurs via a reaction of chlorinate substance with atomic hydrogen that forms at the cathodes and is the main reduction mechanism at hydrogen formation potentials. Additionally, electrochemical oxidation of COCs can be achieved directly at the anode (e.g. boron doped diamond electrodes) [35] or via electrochemically formed hydroxyl radicals and other reactive oxygen species in bulk [29, 30, 36, 37].

The sequential oxidation and reduction processes can be utilized for the transformation of COCs and lead to the creation of less toxic by-products [27, 34, 38]. Authors previously reported that shifts between oxidation and reduction, via electrode polarity reversal, improve TCE removal [39]. However, due to the highly oxidized nature of TCE, inducing HDC as a primary mechanism is valuable and can be supported by placing the cathode upstream of the anode [40].

Multiple reaction zones in the undivided electrochemical cells can be also achieved by the bipolar electrode mode [41]. Bipolar electrolysis is an efficient approach for electrolytes of low electrical conductivity such as groundwater, and it is extensively used for the electrocoagulation/flotation applications. The bipolar mode consists of two feed electrodes at both ends that are connected to a power source and other electrodes without connections. Due to the potential difference between the electrolyte and the electrode, one side of bipolar electrodes becomes anodic, and cathodic polarity generates on the other. Besides iron or aluminum plates, bipolar electrodes, which produce reducing conditions, can consist of zero-valent iron (as packed between feed electrodes) [41, 42]. Also, granular graphite packed-bed bipolar electrodes can be used in order to generate oxidizing conditions [43, 44]. These packed-bed flow-through reactors are efficient due to high active area but corrosion and clogging cause the need for continuous reactor maintenance.

Graphite bipolar electrodes would be the best choice for groundwater treatment since they are not costly and are environmentally friendly (made of non-hazardous materials). However, graphite electrodes are moderately efficient towards both reduction [45] and oxidation [46] when used in monopolar mode. The slow HDC on graphite cathodes is caused by the slow evolution of atomic hydrogen that reacts with the chlorinated chemical. The graphite anodes have limited activity and are defined as modified active anodes; the

oxygen atom to be transferred to an oxidizable substrate first becomes bonded to the previously functionalized anode surface.

Here, we evaluated the use of bipolar plate graphite electrodes to remove TCE from synthetic groundwater. We used a cathode followed by an anode arrangement in an undivided electrochemical cell as novel approach of utilizing graphite material to generate reaction zones for TCE degradation in aqueous solution. We tested the influence of the number of the bipolar electrodes, current intensity and initial TCE concentration on TCE degradation rate.

2. Materials and Methods

All chemicals used in this study were analytical grade. TCE (99.5%) and cis-dichloroethylene (cis-DCE, 97%) were purchased from Sigma-Aldrich. H_2O_2 (30%) was purchased from Fisher Sci. Calcium sulfate was purchased from JT Baker, oxalic acid (anhydrous, 98%) from Acros Organics, sodium chloride, sodium acetate and sodium bicarbonate from Fisher Scientific. Graphite electrodes with dimensions of 3.6 cm diameter by 5 mm thickness were used as electrode materials in all experiments. Deionized water ($18.2 \text{ M}\Omega\cdot\text{cm}$) obtained from a Millipore Milli-Q system was used in all the experiments.

TCE concentration was measured by a 1200 Infinity Series HPLC (Agilent) equipped with a 1260 DAD detector and a Thermo ODS Hypersil C18 column ($4.6 \times 50 \text{ mm}$). The mobile phase was a mixture of acetonitrile and water (60:40, v/v) at 1 mL min^{-1} . 2 mL samples were collected from the sampling ports for analysis.

Analyses of chloride ions were performed using an ion chromatography (IC) instrument (Dionex 5000) equipped with an AS20 analytical column. A KOH solution (35 mM) was used as a mobile phase at a flow rate of 1.0 mL min^{-1} . pH and oxidation-reduction potential (ORP) of the electrolyte were measured by pH meter and ORP meter with corresponding microprobes (Microelectro, USA). The microprobes allow the measurement on these parameters using a small amount of liquid ($\approx 0.2 \text{ mL}$). The amount of H_2O_2 produced during the treatment with bipolar electrodes was analyzed at 405 nm after coloration with TiSO_4 . The measurement was performed at UV-VIS Spectrophotometer (Shimadzu UV1800). The electrochemical reactor with electrode and sampling port locations are shown in Figure 1.

Synthetic groundwater was prepared by dissolving 413 mg L^{-1} sodium bicarbonate and 172 mg L^{-1} calcium sulfate in deionized water. The concentrations of bicarbonate ions and calcium ions are representative of groundwater from limestone aquifers, resulting in electrical conductivity of 800 to $920 \mu\text{S cm}^{-1}$. Excess TCE was dissolved into $18.2 \text{ M}\Omega\cdot\text{cm}$ high-purity water to form a TCE saturated solution (1.07 mg mL^{-1} at 20°C), which was used as stock solution for preparing aqueous TCE solutions. The feedstock solution was stored in a common Tedlar® bag. The headspace in the bag was minimized to limit TCE losses to the gas phase. The initial pH of the contaminated synthetic groundwater was 8 ± 0.3 , and the initial ORP value was $210 \pm 5 \text{ mV}$. The temperature was kept constant at 20°C . Darcy's velocities were selected as 0.25 cm min^{-1} (3 mL min^{-1}). A constant flow velocity was maintained by a peristaltic pump (Cole Parmer, Masterflex C/L). A constant current

intensity during treatments was applied by an Agilent E3612A DC power supply. The experiments conditions are given in Table 1. All experiments were conducted for 180 minutes, and electrolysis steady state conditions were assumed when change in concentration was less 0.5% per minute which accounts for other TCE losses, such as adsorption or volatilization.

TCE removal was calculated by the following equation:

$$\%removal = \frac{c_o - c_t}{c_o} * 100 \quad (\text{Eq. 1})$$

where c_o is the initial TCE concentration (mg L^{-1}) and c_t is TCE concentration at a defined time during treatment (mg L^{-1}). The current efficiency (CE) was calculated according to Faraday's law.

It is important to be able to predict the rate at which contaminants are removed in order to design the full scale application of the technology. The removal kinetic with bipolar mode found to be pseudo-second order (PSO), which is described by Eq. 2 [47, 48]:

$$\frac{t}{c_t} = \frac{1}{c_s} t + \frac{1}{k c_s^2} \quad (\text{Eq. 2})$$

where t is time (min), c_s is the concentration at steady state (mg L^{-1}) and k is PSO coefficient ($\text{L min}^{-1} \text{mg}^{-1}$). Plot of t/c_t against t should give a linear relationship; to confirm the suggested kinetic model, the PSO yields were compared with pseudo-first order (PFO) coefficients.

3. Results and discussion

The influence of the number of bipolar electrodes on TCE removal rate is shown in Figure 2a. The overall TCE removal achieved by monopolar mode was 29% while by bipolar mode with one and two electrodes removal was 38% and 66%, respectively. Results indicate that TCE concentration decay during treatment is affected by the presence of bipolar electrodes. Figure 2b shows the removal kinetics described by the PSO. The correlation coefficients for the PFO kinetic model are low ($R^2 < 0.90$) while there is a good correlation coefficient for the PSO kinetic ($R^2 > 0.98$). PSO coefficients achieved by monopolar mode, bipolar mode with one and bipolar mode two electrodes were ($\text{L min}^{-1} \text{mg}^{-1}$): 0.00855, 0.00936 and 0.177, respectively. The two bipolar electrodes significantly improve the removal rate.

To understand the mechanism of TCE removal we measured the formation of chlorides during the treatment. Chlorides release indicates the reductive HDC process. The chloride yield was low during the application of two bipolar electrodes, which indicates the chlorides oxidation on graphite anodes; the chlorides oxidize to the active chlorine species at the graphite anode [49, 50]. The active chlorine species are strong oxidants, but we assessed that their influence in our study would be insignificant since the amount of chlorides theoretically available from degraded TCE would not generate significant amount of active chlorine species to influence its removal.

Formation of H_2O_2 at different sampling ports during the treatment with two bipolar electrodes is shown in Figure 3. In the undivided electrochemical setups, the products of water electrolysis are competing with the desired reactions on the electrodes; oxygen can be reduced at the cathode to produce H_2O_2 or H_2O [51, 52]. The formation of H_2O_2 on the graphite cathodes follows the equation:

In the cathode followed by anode arrangement in the undivided cell used in our study, the O_2 and H^+ are available as products of the water oxidation at the anodic part of bipolar electrodes (Figure 1). Decomposition of peroxide at the anode is limited since water is in excess, and the decomposition potential between water and peroxide is similar (around 1 V) [53]. Slow decomposition of peroxide at the cathode leads to the formation of hydroxyl radicals through the following reaction:

Hydroxyl radicals ($\text{OH}^\cdot$) oxidize TCE and the degradation byproducts but their activity can be influenced by the presence of carbonates in the groundwater. Carbonate radicals are formed from the reaction of bicarbonate/carbonate with $\text{OH}^\cdot$ and can compromise the oxidation mechanisms induced by $\text{OH}^\cdot$. However, the reaction between carbonate radicals peroxide leads to formation of superoxide which is also a potent oxidizing agent [54].

The production of peroxide at the Port 1 during the first 80 minutes is low, and it is not expected to influence the TCE removal. However, there is increase in the peroxide content at Port 1 up to 2.0 mg L^{-1} . This proves that the peroxide forms as the product of oxygen reduction at the feeder cathode. The shorter inter-electrode distance with the two bipolar electrodes setup than in the monopolar mode allows oxygen formed at the anode part of the bipolar electrode to induce the formation of peroxide at the feeder cathode (Figure 1, Table 1).

The amount of peroxide generated at Port 1 and Port 4 differs: an increase at Port 4 indicates that the processes at the bipolar electrodes contribute to the peroxide formation. The overall the peroxide content after 180 minutes is 36% higher at Port 4 than Port 1.

To confirm the removal mechanism of TCE in the bipolar mode with two electrodes, we measured the TCE decay at each sampling port and data is shown in Figure 4. In order to understand the variations in TCE concentration and mechanisms involved at each sampling port, we compared the results after each 40 minutes of treatment - time is sufficient for the solution to pass through each reaction zone within the reactor.

The removed amount of TCE at Port 4 comparing to Port 1 during the treatment differs indicating different mechanisms within the reactor. Up to 53% of TCE was removed during 80 minutes of treatment after Port 1. The amount of peroxide formed at Port 1 during 80 minutes is not significant (Figure 3) and is less likely contributes to the removal. During further course of treatment the removal of TCE raises up to 61%. The TCE concentration decay is the most extensive after Port 1, which indicates that HDC is the main mechanism of

removal. However, the formation of peroxide that intensifies after 80 minutes of treatment can influence the removal mechanism and rate by creating desirable conditions to oxidize TCE by hydroxyl radicals/superoxide. Yet, the changes in TCE removal at Port 1 from 80 minutes to 120 minutes of treatment were only 4%. Most likely peroxide formation and decomposition on the cathode compromises TCE removal via HDC while concentrations are low for the contribution via oxidation processes.

The contribution of bipolar electrodes (Port 4) to TCE removal increases from 4% after 80 minutes up to 21% after 120 minutes (Figure 3). The results show that bipolar electrodes contribute to TCE removal through the reaction with peroxide decomposition products. The low effect of bipolar electrodes in first 80 minutes of treatment indicates that HDC at cathode surfaces of bipolar electrodes is limited. Direct oxidation and reduction at the electrodes are limited in the flow-through reactors because of the low TCE mass transfer. The contribution of the oxidation at anode is less than 3%.

The influence of current intensity on TCE removal in the system with 2 bipolar electrodes is shown in Figure 5. It is evident that under 120 mA the removal and rate increased compared to 60 mA current intensity; 75% of TCE is degraded under 120 mA after 60 minutes of treatment while 66% was removed under current intensity of 60 mA after 180 minutes. The current efficiency (CE) achieved for 60 mA was 94% while for 120 mA was 73%. The maximum TCE removal achieved was 75% even when the charge amount increased (under 120 mA during 180 minutes). This indicates that the limiting factor for TCE removal in flow-through systems is the mass transfer and proves the pseudo-second order reaction. Also, under the higher current intensities, the bubbles form at the surface of the electrodes and lower the active surface area [55]. The bubbles coverage of the cathode surface adversely affects HDC and can cause the decrease in overall TCE removal.

The removal percentages achieved for 1, 5, 10 and 20 mg L⁻¹ of TCE initial concentration were nearly the same (approx. 70%). Mass flux (j_m , g s⁻¹ m⁻²) towards the cathode for different initial concentrations was: 5×10^{-2} ; 2.5×10^{-1} ; 5×10^{-1} and 1. The data show (Figure 6) that TCE mass removal differs for different initial concentrations (and mass fluxes); with the concentration increase, the availability of TCE molecules for the reactions increases. However, when the same flux was achieved (5×10^{-1} g s⁻¹ m⁻²) by increasing the flow rate (from 3 ml min⁻¹ to 6 mL min⁻¹ with 5 mg L⁻¹ TCE concentration) and by increasing the initial concentration (3 ml min⁻¹ with 10 mg L⁻¹ TCE concentration) different efficiencies were achieved. The increase in flow rate increases the mass flux but lowers the time for interaction of TCE molecules with reactive species/electrode surface.

4. Conclusions

We tested the use of the bipolar electrodes to enhance electrochemical degradation of TCE. The graphite cathode followed by graphite anode sequence operated in monopolar mode (without a bipolar electrode) as well as in bipolar mode with one and two bipolar graphite electrodes in the flow-through electrochemical reactor. The system without bipolar electrodes degraded 29% of TCE while the system with one bipolar electrode degraded 38% of TCE and with two bipolar electrodes 66% of TCE. It was found that the removal

mechanism for TCE degradation in bipolar mode include HDC at the feeder cathode, and oxidation through reaction with peroxide. We conclude that graphite bipolar electrodes enhance TCE removal and can be used to provide simple and practical improvement of electrochemical treatment of groundwater remediation.

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Highlights

Bipolar electrodes enhance electrochemical degradation of trichloroethylene.

Bipolar electrodes generate reduction and oxidation zones to degrade trichloroethylene.

Degradation includes hydrodechlorination at the cathode followed by oxidation with peroxide.

Figure 1.

A schematic of the electrochemical flow-through reactor, the polarization of the bipolar electrodes and example of the reactions occurring at the electrodes. *E1* and *E4* are feeder electrodes; *E2* and *E3* are bipolar electrodes (if used); *P0*, *P1*, *P2*, *P3* and *P4* are sampling ports

Figure 2.

a) The influence of the number of bipolar electrodes and reaction zones on TCE concentration decay (conditions: 5 mg L⁻¹ TCE; 60 mA current intensity; 3 mL min⁻¹ flow) and b) the pseudo-second order rate order plots.

Figure 3.
 H_2O_2 generation during the control treatment (conditions: no TCE; 60 mA current intensity;
 3 mL min^{-1} flow; 2 bipolar electrodes)

Figure 4.

TCE decay at different sampling ports during treatment (conditions: 5 mg L⁻¹ TCE; 60 mA current intensity; 3 mL min⁻¹ flow; 2 bipolar electrodes)

Figure 5.
The influence of current intensity on TCE concentration decay ports during treatment
(conditions: 5 mg L⁻¹ TCE; 3 mL min⁻¹ flow; 2 bipolar electrodes)

Figure 6.

The removal of TCE using different initial concentrations (conditions: 120 mA current intensity; 3 mL min⁻¹ flow; 2 bipolar electrodes)

Table 1Experimental conditions (graphite electrodes and 3 mL min⁻¹ flow)

Mode	Electrode arrangement	Current intensity (mA)	TCE concentration (mg L ⁻¹)
Monopolar	E1→E4	60	5
Bipolar (one bipolar electrode)	E1→E3→E4	60	5
Bipolar (two bipolar electrodes)	E1→E2→E3→E4	60	5
	E1→E2→E3→E4	90	5
	E1→E2→E3→E4	120	5
	E1→E2→E3→E4	60	1
	E1→E2→E3→E4	60	10
	E1→E2→E3→E4	60	20