Original Article

Investigation of Electrocoagulation/Electroflotation Process Efficiency with Aluminum-Graphite felt Electrodes in Removal of E.coli and S. typhimurium from Drinking Water

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Abstract

Introduction:
Electrocoagulation is an electrochemical method for the treatment of water. The present study aimed to investigate the removal of E. coli and S. typhimurium bacteria from drinking water by using Electrocoagulation (EC) - Electroflotation (EF) with Aluminum- Graphite felt electrodes parallel with the monopole mode.

Materials & Methods:
Independent variables included different concentrations of E.coli and S. typhimurium bacteria (104, 105 and 106 CFU/mL), reaction time (5, 10, 15 and 20 min), initial pH (7, 8 and 9), inter-electrode distance (1, 2 and 3 cm), current density (0.83, 1.67 and 2.5 mA/cm2) to determine the optimum conditions.

Results:
The results showed that under optimum conditions the increase in pH from 7 to 9 significantly decreased removal efficiency of bacterial strains of E.coli and S. typhimurium from 100% to 83% and from 100% to 90%, respectively. For the initial concentration of 105 CFU/mL, optimum conditions were obtained 2.5 mA/cm2 for current density, 20 min for reaction time and 2 cm for inter-electrode distance.

Conclusion:
According to the results, efficiency of E.coli and S. typhimurium removal was 100% under optimum conditions. Thus EC/EF process can be used for the removal of pathogenic bacteria from drinking water.

Keywords: Electrocoagulation, Electroflotation, E. coli, S. typhimurium, Drinking Water, Disinfectin

Introduction

Today, many countries in the world are facing difficulties with supplying drinking water, and this situation is worse in developing countries. According to the World Health Organization report, approximately 780 million people were

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deprived of access to drinking water resources in 2012 (1). Moreover, more than 4500 deaths daily occur across the world among children under the age of 14 due to water-associated diseases (2), and 3.4 million people annually die of diseases caused by water in developing countries (3).

The digestive system is a prone part of the body to pathogens, including Escherichia coli (E. coli) as a common pathogen in this organ, which causes traveler's diarrhea and diarrhea in children and adults. E. coli is a gram-negative bacillus from the Enterobacteriaceae family, which is commonly found in the intestine of warm-blooded organisms. E. coli enters the body through water, food, vegetables and fruits, and enters the small intestine using a pathogenic colonization agent that has antigenic properties. These bacteria affect the intestinal osmotic conditions and cause diarrhea by producing resistant and heat-sensitive enterotoxins (4). In developing countries, this agent causes diarrhea in children under the age of 2 (an infection dose of 10^6-10^8 bacteria) and rarely in adults (an infection dose of 10^8 bacteria), whereas in developed countries, it causes diarrhea in adults too (5).

An important serotype of Salmonella enterica is Salmonella typhimurium, whose prevalence of hosting is high and is frequently separated from animal species such as cattle, sheep, goats and poultry. These bacteria are the main cause of food poisoning in humans (6). Salmonella can cause diseases such as gastroenteritis, typhoid or parathyroid fever and septicemia in humans (an infection dose of 10^6-10^8 bacteria). Gastrointestinal infections are the most common presentation of Salmonella-associated infections. The bacteria dominating these infections include Salmonella typhimurium and Salmonella enteritidis. The symptoms of these diseases include nausea, headache, vomiting, acute diarrhea together with a few fecal leukocytes (7-8).

Disinfection of drinking water is a necessity for preventing contagious diseases. A proper disinfectant should have features such as fast and extensive antibacterial properties at ambient temperature, inexpensiveness, not producing byproducts which are harmful to health during usage and aftermath, non-corrosiveness and high solubility in water (9). The most common methods of eliminating pathogenic microorganisms include physical and chemical disinfection of water, separation by membrane (10), heating and using ultraviolet ray, chlorine and its derivatives as well as ozonation (11). Chlorine and its derivatives are normally used in most water treatment plants for disinfection. Although chlorination is a reliable, well-accepted and advanced method, the most important concerns of experts and the public with using this method include producing trihalomethanes, an unpleasant taste and smell and especially risks to human health (12). Furthermore, researchers have shown that, during the disinfection process, different types of byproducts are produced from the reaction between chlorine and natural organic compounds, including haloacetic acids, haloketones, nitrosodimethylamines and haloacet-onitriles (12-13), some of which probably cause cancers including colorectal and bladder cancers (3). Skin reaction to the byproducts during bathing and swimming may also cause carcinogenicity (13). Applying an alternative highly-efficient method for eliminating pathogenic microbes without producing disinfection byproducts therefore appears crucial. Electrochemical methods have successfully been used in recent decades in developed countries to eliminate different contaminants while preserving the environment quality and promoting communities' health. This method is considered an advanced oxidation process in which chemical changes occur as a result of electron transfer at the common surface between the electrode and the solution. The main mechanisms of eliminating microorganisms by electrochemical processes include coagulation, decomposition, reduction, absorption, oxidation, sedimentation and flotation (14). Given that electrochemical processes use different mechanisms to eliminate microorganisms, different phrases are used to refer to them, including electrolytic disinfection, electrochemical disinfection, electrolysis, activated water and anodic oxidation (11 and 14). The present study uses electrolysis to refer to electrical coagulation and electrical flotation. The advantages of this method include simple use, simple equipment, being environmentally friendly, adaptability, safety, selectivity, cost-effectiveness, less sludge production, producing water with less TDS and eliminating the smallest colloidal particles compared to chemical processes (14-15). In addition, there is no possibility of secondary contamination in this method owing to not using chemicals and no need for neutralizing the added chemicals. Solar generators can obviously be used in this process in regions where there is no access to electricity (15).

Zhu et al. who conducted a study in 2005 on eliminating the microbial contamination using the electrolysis process, steel electrodes, felt graphite and an electric current of 0.05-0.25 A achieved an elimination efficiency of 99.99% (16).

Ghernaout et al. conducted a study in 2008 on the efficiency of electrolysis in eliminating E. coli strains using three types of electrodes. They found that aluminum electrode presents the highest efficiency in the destruction of E. coli cells compared to stainless steel and conventional steel electrodes (17).

In 2008, Kraft investigated the elimination of different bacterial strains using the electrochemical method and showed that this method is a proper alternative for the disinfection of cooling water against E. coli, Legionella pneumophila, Salmonella typhimurium and Pseudomonas aeruginosa (11).

The type and configuration of electrodes, the pH of the reactor medium, electric current intensity, initial concentration of the contaminant and duration of reaction are the most important factors affecting the efficiency of the electrical coagulation process. Changing each of these factors can change the nature of electrochemical reactions, energy consumption costs and ultimately the elimination efficiency (18).

The oxygen-based disinfection process is highly reliable in disinfecting drinking water, industrial cooling water and pool water without producing harmful byproducts, which is an advantage of electrochemical-based water disinfection (19). In the electrochemical method, hydrogen peroxide and ozone are produced in the presence of oxygen molecules and electric current. Moreover, the presence of chlorine ions in the solution causes free chlorine and chlorine dioxide to be produced. The main product in the anode is oxygen and chlorine, which is associated with the acidification of the water surrounding the anode. The main product in the cathode is hydrogen, which is associated with alkalizing the water surrounding the cathode and producing calcium carbonate and magnesium hydroxide (11 and 20).

Carbon electrodes, including felt graphite, carbon fibers, carbon electrode and lattice carbon, can cause the production of hydrogen peroxide (21). Owing to its porosity and having many accessible pores, felt graphite has a very large special surface. In fact, this large special surface is a result of thin fibers of the external surface (8-10 microns) and the penetrability of current in the large surfaces between the fibers that can produce free currents from the solution. Moreover, oxygen storage capacity is very high inside the felt graphite plates, and the reaction of free electrons produced in the anode during oxidation can produce OH radical species, ozone, ionic oxygen and other forms of oxygen with bactericidal properties (22). The method used in the present study for eliminating the bacterial strains of E. coli and Salmonella typhimurium was a combination of electrical coagulation and flotation using aluminum and felt graphite electrodes. The general mechanism of electrical coagulation and flotation is a combination of different and simultaneous functions that intensify the function of one another in a chain fashion. The principle mechanism in this process may act through dynamic processes as progressive reactions and change by the type of contaminants as well as environmental and operational factors. If aluminum or ferro is used as the anode, existing contaminants, as a ligand, form polymeric compounds through combining with the ferro or aluminum ions released into the solution. With a very large surface area, these compounds can be very active upon formation in absorbing and compacting contaminants, and given the pH of the solution, they may appear as monomeric or polymeric metal hydroxide ions. Oxygen bubbles that are naturally produced on the anode surface are strong oxidizers and can oxidize organic molecules and turn them to smaller particles (23-24).

Given the water pollution dilemma caused by microorganisms, numerous limitations of conventional water disinfection methods and multiple advantages of the electrolysis process as an applicable process in the consumption point, the present study was conducted to determine the efficiency of the electrolysis process in purifying drinking water contaminated with the strains of E. coli, i.e. indices of microbial contamination of drinking water, and Salmonella typhimurium using aluminum and felt graphite electrodes and a parallel unipolar configuration (25).

Methods and Materials

Chemicals and the waterbed

The present empirical study was conducted in September 2016 in the Microbiology Laboratory of the School of Public Health in Ardabil University of Medical Sciences, Ardabil, Iran using a discontinuous electrochemical reactor in laboratory scale. Sodium chloride was used to adjust electrical conductivity and caustic soda and 1-N hydrochloric acid were used to adjust pH. Table 1 presents the physical and chemical characteristics of the study water solution, which was sterilized before the test. After sterilization, different concentrations of the bacterial strains of E. coli ATCC 25922 and Salmonella typhimurium ATCC 19430 were used to artificially create contamination.

Table 1: The physical and chemical characteristics of the distribution network water of Ardabil

Parameter	Unit	Concentration
Total hardness	mg/L $CaCO_3$	277.55
Calcium hardness	mg/L $CaCO_3$	131.775
Total alkalinity	mg/L $CaCO_3$	225.5
Sodium	mg/L	173.38
Potassium	mg/L	19.07
Nitrate	mg/L	6.01
Nitrite	mg/L	0.22
Sulfate	mg/L	67.02
Chloride	mg/L	96.23
Fluoride	mg/L	0.80
Turbidity	NTU	0.46
Electrical conductivity	mS/cm	0.75
TDS	mg/L	576.45
pH	-----	7.53
Temperature	ºC	18

Preparing the bacterial strains

The bacterial strains used in the present study comprised gram-negative bacteria, including E. coli ATCC 25922 and Salmonella typhimurium ATCC 19430, purchased from the Iranian Industrial Research Center. The bacterial strains of E. coli and Salmonella typhimurium were incubated in the culture medium of Nutrient Broth for 24 hours at 37 ºC in the aerobic condition. A sterile loop was then used for the removal from the culture medium and every bacterial strain was cultured in a uniform linear fashion on specific culture media. i.e. the solid culture medium of MacConkey for E. coli and the solid culture medium of Xylose lysine deoxycholate agar for Salmonella typhimurium. The media were then incubated while inverted for 24 hours at 37 ºC in aerobic conditions (26). All the culture media used had been made by Merck KGaA, Darmstadt, Germany.

Preparing the 0.5 McFarland standard

The McFarland standard was used in the present research as a reference for matching the turbidity caused by the bacterial suspension (27). Given that the number of inoculated bacteria is an important variable affecting the study results, the density of the inoculated microbial suspension should be standard. The optical absorbance of the created turbidity was measured at a wavelength of 610 nm using a 0.5 McFarland solution, including acid sulfuric and chlorobarium, and a spectrophotometer UV/VIS (Model Hach/ DR6000, US), and the optical absorbance was found to be 0.09-0.1. Some bacterial colonies were added to water to create an equivalent turbidity of that measured in the 0.5 McFarland standard tube. Given that the bacterial concentration of 0.5 McFarland is 1.5×108 bacteria/mL, dilution was used to obtain other dilutions (104, 105 and 106 CFU/mL).

Design and construction of the reactor

To conduct the experiments, a reactor with the following specifications was used. The chamber was made of Plexiglas with dimensions of 170×105×135 mm. Two aluminum electrodes and two felt graphite electrodes, each having a dimension of 100×40×2 mm, were alternately connected in a parallel unipolar fashion to a direct current power supply. Distance between electrodes and the floor was 2 cm and distance between electrodes was 1-3 cm. The aluminum electrode was connected to the positive pole of the power supply and the felt graphite electrode to the negative pole. The direct current power supply was of Dazheng DC Power Supply PS-302D type with the capability of generating up to 30 volts. To create mixing inside the reactor during the process, a magnetic stirrer was used with a speed of 200 rpm, which was considered constant in all the experiments. Figure 1 shows the general schematic of the electrical coagulation reactor, the details of the unipolar configuration and the distribution of electrical charges on the electrodes (28).

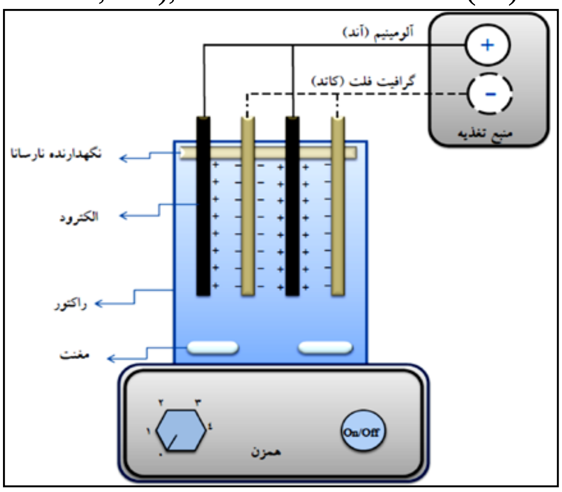

Figure 1: Electrochemical reactor and the parallel unipolar connection of electrodes to power supply

Chemical and biological tests

The electrodes were made of commercial aluminum and felt graphite plates. The electrodes surfaces were rubbed with a sandpaper and immersed in an HCL solution and then rinsed in distilled water for one minute to remove impurities from the electrodes before conducting each test. The present study investigated the effects of five independent variables, including current density, duration of reaction, distance between electrodes, initial pH and initial bacterial concentration, to determine the efficiency of the electrolysis process in removing gram-negative bacteria such as E. coli and Salmonella typhimurium from drinking water. Every test began with adding 1.5 liters of water with a bacterial concentration of 105 bacteria/mL and a pH of 7, 8 and 9 to the reactor and the electrolysis was performed using current densities of 0.83, 1.67 and 2.5 mA/cm2 and a distance between electrodes of 1, 2 and 3 cm. After 5, 10, 15 and 20 minutes, the content of the reactor was sampled to determine the degree of bacterial elimination. An optimal operation parameter was defined in every test and the process efficiency was ultimately determined for the given current density, distance between electrodes, duration of reaction, optimal pH and initial concentrations of 104, 105 and 106 bacteria/mL.

Given that the tests on the bacterial strains of E. coli and Salmonella typhimurium were separately conducted, 288 samples were obtained through three repetitions of the tests. Through three iterations, the number of samples was calculated as 48 for determining the optimal electrical conductivity and controlling the solution temperature.

All these tests were conducted at the laboratory temperature of 20±2 ºC. Measurements of pH, voltage and temperature were made during the tests. A pH meter (Model Hack, US) was used to measure pH, an electric conductivity meter (Model Hack, US) to measure electrical conductivity and a mercury thermometer to measure the solution temperature.

Approximately 5 mL of the electrolyzed solutions was removed as a sample and kept in sterile culture tubes. After 30 minutes of sedimentation, part of the supernatant was removed and the necessary dilation was carried out. One hundred µL of the diluted solution was then cultured on the specific agar culture medium of every bacterial strain. Equation 1 is used to calculate the efficiency of bacterial elimination.

$$E\% = \left(\frac{C_{in} - C_{out}}{C_{in}}\right) \times 100 \qquad (1)$$

Where E% represents the percentage of bacterial elimination, Cin the number of input colonies and Cout the number of output colonies following the electrolysis process. All the tests were separately conducted for the bacterial strains of E. coli and Salmonella typhimurium as per the existing guidelines in the book Standard methods for the examination of water and wastewater (26). It is worth noting that the range of pH selected was according to the pH values reported for drinking, surface and underground water and it is adapted to water treatment plants, in which the pH of the water treated is usually close to 7. To determine the amount of electrode consumed for refining every liter of drinking water, an accurate scale was used to measure the weight of the electrodes at the beginning and the end of the process and the difference between the electrodes' weights was considered the consumed electrode. Equation 2 was also used to calculate energy consumption during the electrolysis process (29):

$$E = \frac{IUT}{V} \qquad (2)$$

Where E is energy consumption (kWh/m3), I current (A), U the voltage across the reactor (V), V the volume of water inside the reactor (m3) and T is the process duration (h).

Statistical analyses

The statistical analyses of the data were conducted in SPSS-14 and the diagrams were drawn in Excel. One-way ANOVA was used to investigate the significance of the relationship between the initial and the remaining concentration of the bacterial strains and to determine the correctness of the process efficiency in removing the bacterial strains.

Results

As shown in diagram 1, the solution temperature increases with an increase in the reaction duration and electrical conductivity. The maximum temperature of the solution was 53 ºC at an electrical conductivity of 13 mS/cm and a contact duration of 20 minutes. In this case, the temperature variation was equal to 30 ºC from the beginning to the end of the reaction. The minimum temperature of the solution was obtained at an electrical conductivity of 0.9 mS/cm with a range of temperature variation of 3 ºC from the beginning to the end of the reaction.

As table 2 suggests, distance between electrodes does not significantly affect the electrolysis process. In fact, a decrease in the number of bacteria was nearly constant and equal to over 98%. Distance between electrodes was therefore considered 2 cm in all the tests. With a 2-cm distance between electrodes and an electrical conductivity of 0.9 mS/cm, the number of bacterial strains of E. coli and Salmonella typhimurium was reduced by 100% (Table 2).

Table 3 presents the effect of pH on reducing the number of bacteria. With an increase in pH from 7 to 9, the efficiency of eliminating the bacterial strains of E. coli decreases from 100% to 83% and Salmonella typhimurium from 100% to 90%. During the optimization stages, the pH variations of the water sample inside the reactor were measured at the beginning and the end of the reaction, as shown in table 3. The mean variations in pH were found to be 0.5.

The effect of variations in current density on the efficiency of eliminating the strains of E. coli and Salmonella typhimurium is shown in diagram 2, suggesting an increase in the efficiency with an increase in current density. In fact, with an increase in current density from 0.83 to 2.5 mA/cm2 and a contact duration of 20 minutes, the efficiency of eliminating the strains of E. coli and Salmonella typhimurium respectively increased from 84% and 89% to 100% in the optimal conditions.

Diagram 3 shows the results obtained from eliminating the bacterial strains of E. coli and Salmonella typhimurium at 5, 10, 15 and 20 minutes. The efficiency of eliminating both bacterial strains increased with an increase in the reaction duration. With an increase in the reaction duration from 5 to 20 minutes, the efficiency of eliminating the strains of E. coli and Salmonella typhimurium respectively increased from 55% and 56% to 100%.

After optimizing all the parameters, the optimal condition was ultimately obtained in terms of efficiency, operation and economy of the process at pH=7, current density=2.5 mA/cm2, distance between electrodes=2 cm, electrical conductivity= 0.9 mS/cm and duration of reaction=20 minutes. According to the determined optimal conditions, the effect of the initial bacterial concentrations was investigated separately, and the results are shown in table 4.

The results of the ANOVA between factors such as the initial concentration of the bacterial strains of E. coli and Salmonella typhimurium and their remaining concentration are shown in table 5. The statistical analysis suggests a significant relationship between the initial bacterial concentrations and the process efficiency ($P<0.01$)

The electrode consumption for purifying every liter of drinking water was calculated in optimal conditions by measuring the variations in the weight of anode (aluminum) and cathode (felt graphite) electrodes at the beginning and the end of the process (Table 6).

Table 7 shows energy consumption in different conditions, including a current density of 0.83, 1.67 and 2.5 mA/cm2, duration of reaction of 5, 10, 15 and 20 minutes and a pH of 7, 8 and 9. The initial concentration of the bacterial strains of E. coli and Salmonella typhimurium was equal to 105 CFU/mL. As shown in table 7 and given the elimination efficiency of the bacterial strains, a current density of 2.5 mA/cm2 and a duration of reaction of 20 min are considered optimal operational conditions.

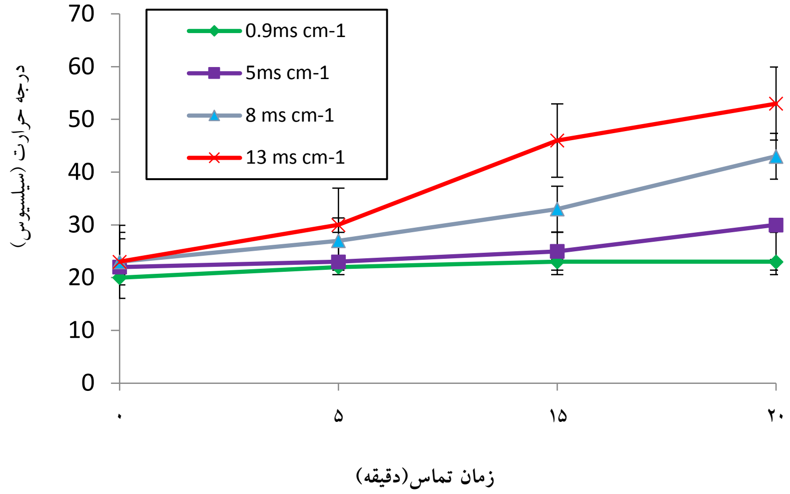

Diagram 1: Variations in temperature with an increase in the contact duration and electrical conductivity during the electrolysis process (Voltage: 30 V)

Table 2: The effect of distance between electrodes on the efficiency of the electrolysis process in removing the bacterial strains (pH=7, electrical conductivity=0.9 mS/cm, initial bacterial concentration=10^5 CFU/mL, current density=2.5 mA/cm^2, duration of reaction=20 min and sedimentation duration=30 min)

Distance between electrodes (cm)	1	2	3
Reduction in the number of E. coli (%)	99.2	100	98
Reduction in the number of Salmonella typhimurium (%)	99.7	100	99

Table 3: The effect of the initial pH on the efficiency of the electrolysis process (current density=2.5 mA/cm^2, electrical conductivity=0.9 mS/cm, duration of reaction=20 minutes, distance between electrodes=2 cm, initial bacterial concentration =10^5 CFU/mL and sedimentation duration=30 min)

Initial pH	7	8	9
Final pH	7.5	8.4	9.6
Reduction in the number of E. coli (%)	100	94	83
Reduction in the number of Salmonella typhimurium (%)	100	97	90

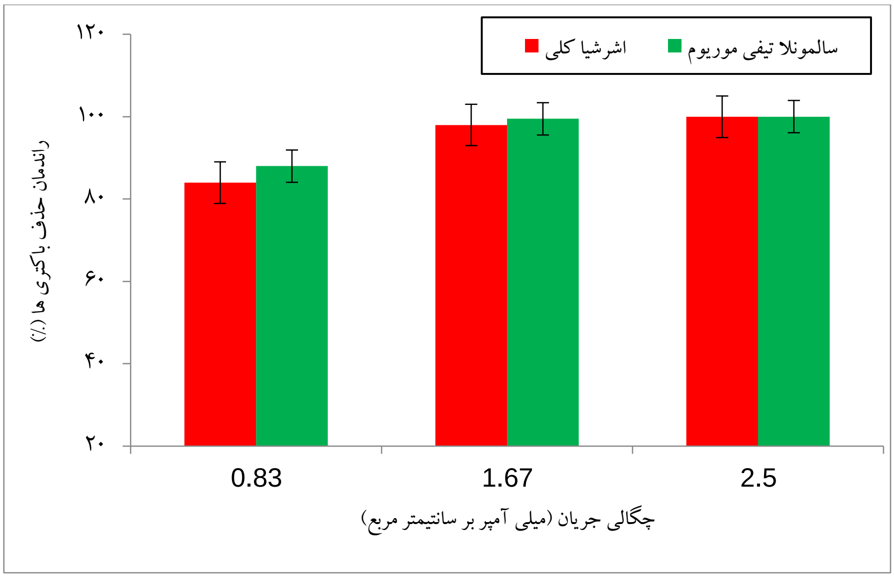

Diagram 2: The effect of current density on the efficiency of the electrolysis process (pH=7, duration of reaction=20 min, distance between electrodes=2 cm, electrical conductivity=0.9 mS/cm, initial bacterial concentration=10^5 CFU/mL and sedimentation duration=30 min)

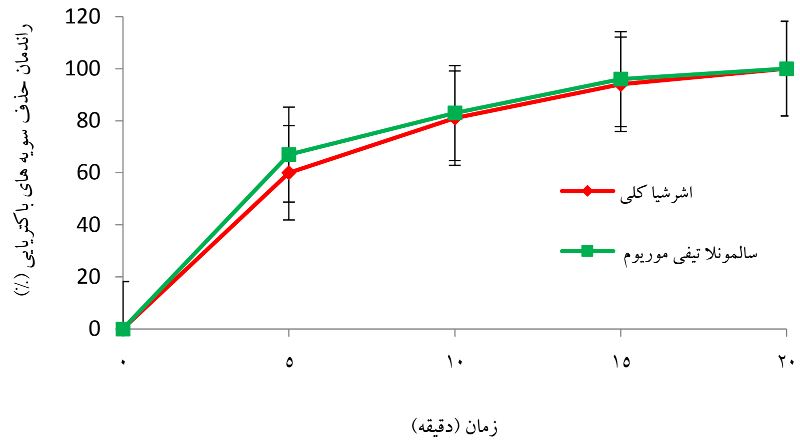

Diagram 3: The effect of contact duration on the efficiency of the electrolysis process (pH=7, current density=2.5 mA/cm^2, distance between electrodes=2 cm, electrical conductivity=0.9 mS/cm, initial bacterial concentration=10^5 CFU/mL and sedimentation duration=30 min)

Table 4: The results of the elimination efficiency (in %) of the bacterial strains at different initial concentrations (pH=7, duration of reaction=20 min, current density=2.5 mA/cm^2, distance between electrodes=2 cm, electrical conductivity=0.9 mS/cm and sedimentation duration=30 min).

Bacterial strain	Parameters of the elimination efficiency	Initial concentrations of the bacterial strains of E. coli and Salmonella typhimurium (CFU/mL)		
		10^4	10^5	10^6
E. coli	Mean	99.76	100	77
	Standard Deviation	0.205	0	1.41
Salmonella typhimurium	Mean	99.93	100	80
	Standard Deviation	0.053	0	1.29

Table 5: The results of the One-way ANOVA between the initial concentration of the bacterial strains and their remaining concentration

Bacterial strain	Initial bacterial concentration	Mean remaining bacterial concentration	Standard deviation	P
E. coli	10^4	25	22.9	0.000
	10^5	0.00	0.00	0.000
	10^6	230000	10000	0.000
Salmonella typhimurium	10^4	13.3	4.04	0.000
	10^5	0.00	0.00	0.000
	10^6	200000	20000	0.000

Table 6: Variations in the weight of the electrodes used in the process

Electrode type	Weight at the beginning of the process (g)	Weight at the end of the process (g)	Electrode consumption (g/L)
Aluminum	21.4520	21.428	0.016
	21.758	21.738	0.013
Felt graphite	5.34	5.3397	0.00023
	4.95	4.9498	0.00019

Table 7: Energy consumption by current density, duration of reaction and pH (the initial concentration of the bacterial strains=10^5 CFU/mL, distance between electrodes=2 cm and the sedimentation duration=30 min)

Current density (mA/cm^2)	Duration of reaction (min)	Energy consumption (kWh/m^3)		
		pH=7	pH=8	pH=9
0.83	5	0.042	0.0453	0.0407
	10	0.0860	0.0926	0.0834
	15	0.1295	0.1394	0.1256
	20	0.168	0.1812	0.1628
1.67	5	0.1282	0.1393	0.1398
	10	0.2584	0.2805	0.2816
	15	0.3886	0.4219	0.4234
	20	0.5128	0.5573	0.5592
2.5	5	0.4538	0.4672	0.5027
	10	0.9106	0.9373	1.0084
	15	1.3674	1.4076	1.5141
	20	1.8172	1.8709	2.0128

Discussion

The effect of electrical conductivity on the solution temperature and electrolysis process

The intensity of electric current flow determines the amount of coagulant and the amount of hydrogen, hydrogen peroxide, ozone and other molecular and ionic species produced (11). Moreover, the current flowing into the solution changes its temperature (30). The effect of temperature on the microbial deactivation has been reported in magnetic fields (31). To control temperature during purification using the electrolysis process, an initial study was conducted to select electrical conductivity. The electrolysis process was performed in a discontinuous fashion using a voltage control of up to 30 volts. As observed in diagram 1, the solution temperature increases with time and electrical conductivity. For a given voltage (ΔV), the electrolysis current (I) depends on the solution resistance (R) between electrodes, i.e. $I= \Delta V/R=\Delta V.G$. An increase in the solution electrical conductivity (G) increases electric current, causing an increase in the solution temperature due to the increased energy consumption (RI2) (Diagram 1). Given that most pathogenic bacteria remain alive at a temperature of 15-45 ºC, the solution temperature was controlled to be below 40 ºC by selecting solutions with a properly low electrical conductivity. On the other hand, given electrical conductivity as a key parameter in the electrolysis process, its value should be high enough to allow the flow of current through the solution. Diagram 1 shows that the lowest variations in the solution temperature from 20 to 23 ºC were obtained for an electrical conductivity of 0.9 mS/cm. With a 2 cm distance between electrodes and an electrical conductivity of 0.9 mS/cm in optimal conditions, the reduction in number of the bacterial strains of E. coli and Salmonella typhimurium reached 100% (Table 2). Electrical conductivity was therefore considered to be 0.9 mS/cm in all the experiments. The reduction observed in the number of bacteria can therefore be only attributed to the electric field applied in the purification process. Ricordel et al. who studied the mechanism and efficiency of reducing the number of E. coli using electrical coagulation and aluminum electrodes in a water medium found that the solution temperature increases with electrical conductivity. To control the temperature, they therefore used solutions with lower electrical conductivities, i.e. below 0.6 mS/cm, and found that the reduction in the number of E. coli was only caused by the applied electric field (32).

Effects of distance between electrodes

With a distance between anode and cathode electrodes of 1, 2 and 3 cm, an electrical field of 30, 15 and 10 V/cm was respectively applied. As table 2 suggests, the elimination efficiency remains nearly constant with an increase in distance between electrodes and the distance does not significantly affect the electrolysis process. In most of the experiments, distance between electrodes was therefore considered 2 cm. The principle mechanism behind the elimination of bacteria by the electrolysis process has not yet been completely understood. Biological cells have been known to show intense physiological responses when exposed to magnetic (or electric?) fields. Most of these responses are based on changes in the potential between the membrane, which is caused by applying an external magnetic (or electric?) field. Applying magnetic (or electric?) fields of higher than a threshold (normally 0.2-1 V) can cause the formation of pores in the membrane, and this phenomenon is called electric field-induced membrane pores (33).

Effects of the solution pH

The solution pH plays a key role in the rate and efficiency of chemical reactions, including the electrolysis process. The solution pH changes during the electrolysis reaction owing to the production of hydrogen in the cathode and the release of dissolved carbon dioxide caused by hydrogen bubbles being mixed. This change in pH depends on the water alkalinity, electrode material and the initial pH of water. The medium pH should be constantly controlled and regulated to control the process efficiency owing to the extreme dependency of coagulation on pH. Research suggests that the effect of pH depends on the type of contaminant and process (32). Table 3 presents the results of the effects of pH on reducing the number of the bacteria. Neutral and slightly alkaline are the best pH for eliminating the bacterial strains of E. coli and Salmonella typhimurium. The elimination efficiency declines with an increase in the medium alkalinity. With an increase in the initial pH from 7 to 9, the elimination efficiency of the bacterial strains of E. coli is significantly reduced from 100% to 83% and that of Salmonella typhimurium from 100% to 90%, which can be explained by the solubility of aluminum in different values of pH. Aluminum ions are mainly found to be Al(OH)3 in pHs of 7 and 8.5, which have a sedimentary state. The bacteria can be absorbed on the active places of these clots and eliminated from the system through sedimentation or flotation. An increased solubility of Al+3 causing a reduction in the produced clots can be another cause of the efficiency decline (34). The optimal pH in the purification process is equal to 7, as the efficiency is maximum in pH=7. The present study therefore selected pH=7 as the optimal condition in terms of the elimination efficiency and purification process operation.

Effects of current density

The electrolysis reaction begins with applying an electric current. Severe current fluctuations affect the amount of coagulant produced and the process efficiency. The electric current changes should therefore be investigated in terms of energy consumption optimization and ultimately the process economy. The proper optimization of current intensity can affect the process efficiency and the economic aspects of the process and reduce the direct costs of electrical energy and indirect costs associated with electrode consumption. Diagram 2 shows the effect of changes in current density on the elimination efficiency of the bacterial strains of E. coli and Salmonella typhimurium. Electric current density was investigated in the following optimization steps as an effective factor in the elimination efficiency. This factor affects the rate of electrolysis processes through influencing the number of metal ions that are removed from the electrode surface (35). To optimize current density at different levels, including 0.83, 1.67 and 2.5 mA/cm2, the optimal distance between electrodes was considered to be 2 cm, the optimal pH to be 7 and the reaction duration to be 20 minutes. Diagram 2 suggests that the lowest elimination efficiency is associated with the lowest current density and equals 84% for the bacterial strains of E. coli and 89% for Salmonella typhimurium. With an increase in current density to 1.67 mA/cm2, the elimination efficiency of E. coli and Salmonella typhimurium significantly increased by 14% and 10.5% respectively. This increase in efficiency can be explained by an increase in the decomposition rate of aluminum electrodes and an increase in the production of metal hydroxides caused by an increase in the current flow through the water sample. Aluminum hydroxide is formed by dissolving metal ions which are separated from the reactant electrode (aluminum), first causing the destabilization of microorganisms and then causing the coagulation of unstable phases and elimination of microorganisms through sedimentation/separation (36). In low current densities, the number of aluminum ions separated from electrodes is low, causing a proportionate drop in the elimination efficiency of the bacteria. The results of the present study on changes in current density and their effect on the elimination efficiency of bacteria are consistent with those obtained in similar studies (17 &33).

As shown in diagram 2, the elimination efficiency of the bacterial strains of both E. coli and Salmonella typhimurium is respectively increased from 98% and 99.5% to 100% with an increase in current density from 1.67 to 2.5 mA/cm2. According to the guideline of the Institute of Standards & Industrial Research of Iran, every 100 mL of all types of drinking water should be negative in terms of index bacteria such as E. coli and thermoplastic coliforms (37). The optimal current density for eliminating the bacterial strains of E. coli and Salmonella typhimurium was therefore considered to be 2.5 mA/cm2.

Effects of duration of reaction

Duration of reaction is an effective factor in the elimination efficiency of bacteria (35). Diagram 3 illustrates the results associated with eliminating the bacterial strains of E. coli and Salmonella typhimurium for reaction durations of 5, 10, 15 and 20 minutes. Duration of reaction has always been studied as an effective factor in all process and operational units in water treatment plants (35). The changes in this factor and their effect on the process efficiency was investigated in the following optimization step. The optimal factors obtained in the previous steps were considered when optimizing the reaction duration; distance between electrodes was considered to be 2 cm, pH=7 and current density=2.5 mA/cm2 for reaction durations of 5, 10, 15 and 20 minutes. With a reaction duration of 5 minutes, the elimination efficiency of the bacterial strains of E. coli was 60% and that of Salmonella typhimurium was 67%. With an increase in reaction duration to 15 minutes, the elimination efficiency of E. coli increased to 94% and that of Salmonella typhimurium to 96%. The efficiency increased to 100% for both types of bacteria with 20 minutes' duration of reaction. According to Faraday's law, duration of reaction directly affects the solubility of the anode metal. With an increase in the duration of reaction, more Al+3 ions are separated from the surface of the aluminum electrode, therefore increasing the concentration of Al+3 and aluminum hydroxide flocks and affecting the electrolysis process efficiency (38). Although increasing the reaction duration increases the electrolysis process efficiency, it increases the costs of electrical energy and electrode consumption. A 20-minute duration of reaction was ultimately selected as the optimal duration.

Effects of the optimized values on different concentrations of bacterial strains

The present study examined the initial concentrations of the bacterial strains of E. coli and Salmonella typhimurium as effective factors. The results shown in table 4 suggest that the elimination efficiency remains nearly constant with an increase in the initial bacterial concentration from 104 to 105 CFU/mL; however, with an increase in the initial concentration from 105 to 106 CFU/mL, the elimination efficiency significantly decreases to 77% for E. coli and 80% for Salmonella typhimurium. An increase in the number of bacteria available in the reactor against the amount of coagulant is the most important reason for this effect on the process efficiency, since different bacterial concentrations enter the reactor at the same medium conditions while the amount of energy consumption and consequently the amount of generated coagulant remain constant. The elimination efficiency of the bacteria reduces with an increase in their initial concentration. Given the values determined for the effective factors in the electrolysis process, a certain amount of hydroxide compounds is generated, which is able to remove only a certain number of microorganisms. These result are consistent with those obtained in similar studies in which the elimination efficiency decreased with an increase in the initial bacterial concentration while an increase in electric current and reaction duration was needed to increase the efficiency (32 and 39). According to the obtained results, the efficiency of bacterial elimination is optimal for the initial concentrations of 104 and 105 CFU/mL, whereas the process efficiency at a concentration of 106 CFU/mL is not convincing and needs more investigations. Research, however, suggests the water entering the disinfection unit of water treatment plants contains fecal coliforms of less than 100 CFU/mL (40-41). The electrolysis process can therefore be used as a disinfection unit to eliminate pathogenic bacteria.

According to table 5, One-way ANOVA was used to investigate the significance of the relationship between the initial concentrations of the bacterial strains and the remaining concentrations and to confirm the process efficiency obtained for eliminating the bacterial strains. The statistical test of the data in table 4 suggests the dependency of the remaining concentration of the bacterial strains on the initial concentration, therefore suggesting a significant relationship between the initial bacterial concentration and the process efficiency. This relationship is significant with a 99% coefficient of confidence (Table 4).

Energy and electrode consumption

As for electrode consumption, a higher weight reduction was observed in anode or the sacrificial electrode (aluminum) compared to the cathode electrode (felt graphite). In other words, the mean electrode consumption for purifying one liter of water was 0.029 for aluminum and 0.00042 for felt graphite. The results indicated no weight changes in the cathode electrode during the process and the associated consumption can therefore be considered zero (Table 6), whereas the main cost of electrode consumption is associated with aluminum, which should be examined when designing water treatment plants. An increase in either current density or duration of reaction was found to increase energy consumption, which is consistent with other studies (42-43). With the optimal conditions for the study process, the electric power consumption equals 1.8172 kwh/m3. Although the process efficiency increases with duration of reaction and current density, the economic costs associated with electrode and energy consumption should be addressed in the optimal condition.

Conclusion

The results obtained from the present study suggest that the elimination efficiency of the bacterial strains of E. coli and Salmonella typhimurium is significantly reduced with an increase in pH, and is significantly increased with an increase in current density and duration of reaction. Furthermore, an increase in the initial concentration of the bacterial strains reduces the process efficiency. Distance between electrodes does not significantly affect the process elimination efficiency. The solution temperature, which was found to increase with an increase in the duration of electrolysis reaction, can be controlled by selecting a proper electrical conductivity. Given the 100% efficiency obtained for eliminating the bacterial strains in the optimal conditions, the electrolysis process can be used to remove pathogenic bacteria from drinking water. In addition, the economic costs associated with this process, including the costs of energy consumption and aluminum electrode consumption, should be considered when selecting the optimal conditions of purification, operation and working efficiency. The method reported in the present research can be used as a reliable, effective, fast and economical approach to eliminating bacterial strains from water media. It will also help resolve most of the problems associated with pathogens in drinking water.

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Conflicts of Interest

None declared.

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