



Comparing the growth and structure of flocs from electrocoagulation and chemical coagulation



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ABSTRACT

This study compares the growth and structure of flocs from chemical coagulation (CC) and electrocoagulation (EC). Flocs were more compact and larger when using CC rather than EC (on average, scattering exponents were 2.60 versus 2.31, while floc sizes were 254 versus 144 μm), and in low rather than high salt (2.51 versus 2.40, 222 versus 181 μm). They were also more compact at final pH 8.3 rather than pH 6.0 (2.53 versus 2.38). Transmission electron microscopy (TEM) revealed that CC and EC flocs were structurally distinct, suggesting that the former is better at aggregating. In low salt and at pH 8.3, flocs were more stable and likely required more collisions to form, producing denser structures. Compact flocs tend to be more resistant to breakage, allowing them to grow to larger sizes. The time required for a floc size distribution to stabilize depended strongly upon the interaction between method of dosing and final pH. The CC-pH 6.0 and EC-pH 8.3 cases stabilized fastest (averaging 8.0 and 7.8 min), as they were always in the appropriate pH range for iron precipitation. The CC-pH 8.3 cases were initially adjusted close to pH 9 to counter coagulant acidity, while the EC-pH 6.0 cases possibly suffered from localized acidity near the anode, making precipitation less successful.

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1. Introduction

1.1. Electrocoagulation

In water treatment, coagulation is a process wherein additives are used to destabilize particles in solution, thus making them more likely to aggregate upon collision [1]. Coagulation can be induced using chemical salts (such as ferric chloride or alum), or via electrocoagulation (EC), which uses sacrificial electrodes to provide a pure source of cations. Unlike chemical coagulation (CC), EC is not a commonly used water treatment technology. Nevertheless, EC has successfully treated a diverse variety of water types at the bench-scale, including municipal, textile dye, and petroleum refinery wastewaters [2–4].

An EC system includes an electrochemical cell; it must contain at least one anode-cathode pair, along with an electrolyte and power source. While hydrogen gas is produced at the cathode surface, metal cations are released at the anode surface and react with water to form a variety of metal-hydroxide complexes. The rate

of cation generation can be controlled by adjusting the applied current, according to Faraday's law (Eq. (1)):

$$m = \frac{ItM}{zF} \quad (1)$$

where m is the mass of metal cation generated (g), I is the current (A), M is the molecular weight of the metal (g/mol), z is the number of electrons transferred per atom, and F is Faraday's constant (96 485 $^\circ\text{C}/\text{mol}$). Other papers describe these electrochemical reactions in greater detail [5,6].

EC has certain advantages over CC. For example, EC typically consumes less alkalinity and requires fewer chemical additives for pH adjustment. Solid metal electrodes are also easier to store and move than corrosive chemical salts. These qualities potentially make EC an option for use in remote areas or in emergency situations [7,8].

1.2. Floc development

Floc growth begins with the aggregation of primary particles (which are simply the "original," unbound particles [1]). Initial floc growth is rapid, forming large, porous structures. As the process continues, floc breakage becomes more prominent. The rates of aggregation and breakage eventually equalize, creating stable particle size distributions [9–11]. Floc growth is affected by a number of

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factors, such as coagulant concentration and applied shear. Higher coagulant doses tend to increase both floc size and growth rate, as there are more particles available for aggregation [9,11]. Furthermore, mixing at greater intensities (higher shear) tends increase breakage, thereby reducing floc size and creating more compact structures [11,12].

1.3. Comparing chemical coagulant and electrocoagulation

A number of papers have compared CC and EC. Perhaps unsurprisingly, their results are widely varied as the experimental systems and/or the solutions under treatment are all unique. For example, Bagga et al. [7] used EC as a pretreatment step for surface water microfiltration, but found that CC with FeCl_3 was better at reducing membrane fouling. They found that EC produced soluble ferrous iron, which was less effective than ferric iron. Zhu et al. [8] experimented with the removal of viruses from a synthetic freshwater using CC or EC, followed by microfiltration. The authors found that EC outperformed CC, and proposed that virus adsorption and/or enmeshment was improved due to localized regions near the anodes having lower pH levels, as well as higher iron and virus concentrations. Cañizares et al. [13] compared EC with aluminum electrodes against CC with alum for the break-up of oil-in-water emulsions. Unlike either Bagga et al. [7] or Zhu et al. [8], they found that the process efficiency depended on the pH and the aluminum concentration, but not the technology itself. As a final example, Harif et al. [14] studied floc formation in a suspension of kaolin, comparing CC with alum and EC with aluminum electrodes. They determined that, when compared to CC flocs, EC flocs were more fragile, but formed faster and over a wider pH range.

Studies regarding EC flocs are limited, particularly in salt water. This paper compares the growth and structure of CC and EC flocs in low and high salt solutions at two pH levels, analyzing differences in zeta potential, scattering exponent (an approximation of fractal dimension), particle size distributions, and transmission electron microscopy (TEM) images. Floc characteristics are relevant to the effectiveness of downstream processes in a water treatment train, such as settling or filtration.

2. Materials and methods

2.1. Bench-scale EC system

The bench-scale EC system used for this experiment is diagrammed in Fig. 1. The system had a working volume of 1170 mL, and was operated in batch. The electrodes were a single anode–cathode pair. The anode was pure iron, and the cathode was stainless steel (SS). The anode was a wire with a 2 mm diameter and a 5.1 cm submerged height, resulting in 3.20 cm^2 of submerged surface area. The cathode was a half-cylinder with a 2.1 cm outer diameter, a 0.25 cm wall thickness, and a 5.1 cm submerged height, resulting in 32.2 cm^2 of submerged surface area. There was a space of 0.7 cm between the anode surface and the inner diameter of the cathode, and the surface area ratio of the cathode to the anode was 10.0. The electrodes were clamped into place using non-conductive plates, and could be easily disassembled for cleaning. The electrodes were scrubbed with steel wool and rinsed with reverse osmosis (RO) water between test runs.

A GPS 3030DD DC power source (GW Instek, New Taipei City, Taiwan) was used to supply current. Current was recorded by placing a 1Ω resistor (1% error tolerance) in series with the EC cell, and then connecting a Volt101A data-logging voltmeter (Madtch, Warner, NH, USA) in parallel with the resistor. By Ohm's law and Kirchhoff's laws, the voltage across the resistor was equivalent to the current of the system. The logged currents generally reached

Table 1
Experimental factors and levels.

Factor	(–)	(+)
Final pH	6.0	8.3
Salt concentration	0.25 g/L NaCl, 0.50 g/L NaHCO_3	25 g/L NaCl, 0.50 g/L NaHCO_3
Method of dosing	Chemical coagulation	Electrocoagulation

some plateau value within a few seconds, and then remained constant with minor fluctuations. For each EC test, the median current was calculated and considered to be the steady-state current.

2.2. Electrolyte

This research varied two electrolyte parameters: the salt concentration, and the final pH. Regarding salt concentration, the low salt electrolyte consisted of 0.25 g/L NaCl and 0.50 g/L NaHCO_3 dissolved together in RO water. This resulted in an ionic strength of 10^{-2} M and an average initial conductivity of 1.12 mS/cm (standard deviation 0.04 mS/cm). The high salt solution used 25 g/L NaCl and 0.50 g/L NaHCO_3 , resulting in an ionic strength of 0.43 M and an average initial conductivity of 42.2 mS/cm (1.6 mS/cm). For comparison, the ionic strength of seawater is approximately 0.7 mol/kg , while the ionic strength of freshwater ranges from 5×10^{-4} to 10^{-2} M [15]. The solution alkalinity was 298 mg/L as CaCO_3 . Note that oceans can have alkalinity levels in the ballpark of $2300 \mu\text{mol/kg}$, or 230 mg/kg as CaCO_3 after conversion [16].

Furthermore, the solutions were pH adjusted such that the final pH after coagulant addition would be either 6.0 or 8.3. pH adjustments were made using dilute solutions of HCl and/or NaOH, as necessary. The average final pH values for each group were 6.00 (standard deviation 0.07) and 8.30 (0.07). All chemicals used in this experiment were purchased from Fisher Scientific (Waltham, MA, USA).

2.3. Experimental design and procedure

The experimental design for this project was a 2^3 factorial design. The experimental factors studied were final pH (6.0 and 8.3), salt concentration (0.25 and 25 g/L NaCl), and method of coagulant dosing (CC and EC). This resulted in 8 combinations, which were run in duplicate. The factors and levels are listed in Table 1.

Each test began by filling a jar with 1170 mL of salt solution. The solution was pH adjusted such that the final pH after CC or EC would target either 6.0 or 8.3. A stir bar was set to continuously mix the solution at a speed of 200 rpm. Initial pH, conductivity, and zeta potential measurements were taken. The particle sizer was primed with the test solution and setup to take measurements once every minute.

Coagulant was added continuously over 5 min using either CC or EC. With the former, 25 mg/L FeCl_3 hexahydrate was dosed from a 5 g/L stock solution at a rate of 1.2 mL/min using a 7543-12 Masterflex pump (Cole-Parmer, Vernon Hills, IL, USA). With the latter, a targeted concentration of 5.2 mg/L of pure iron was generated from a sacrificial anode using 7.3 V and 1.3 V for the low and high salt concentrations, respectively. The relationship between voltage and per-minute iron output was established experimentally before the start of the tests for both the low salt (Eq. (2)) and high salt (Eq. (3)) solutions:

$$F = 0.171 V - 0.201R^2 = 0.903 \quad (2)$$

$$F = 3.087 V - 2.947R^2 = 0.978 \quad (3)$$

where F is the iron concentration (mg/L), and V is the operating voltage (V). Working backwards from Faraday's law (Eq. (1)), the targeted current was 0.088 A , assuming a z -value of 2.5 (in other

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