



General Potential-Current Model and Validation for Electrocoagulation



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ABSTRACT

A model relating potential and current in continuous parallel plate iron electrocoagulation (EC) was developed for application in drinking water treatment. The general model can be applied to any EC parallel plate system relying only on geometric and tabulated input variables without the need of system-specific experimentally derived constants. For the theoretical model, the anode and cathode were vertically divided into n equipotential segments in a single pass, upflow, and adiabatic EC reactor. Potential and energy balances were simultaneously solved at each vertical segment, which included the contribution of ionic concentrations, solution temperature and conductivity, cathodic hydrogen flux, and gas/liquid ratio. We experimentally validated the numerical model with a vertical upflow EC reactor using a 24 cm height 99.99% pure iron anode divided into twelve 2 cm segments. Individual experimental currents from each segment were summed to determine total current, and compared with the theoretically derived value. Several key variables were studied to determine their impact on model accuracy: solute type, solute concentration, current density, flow rate, inter-electrode gap, and electrode surface condition. Model results were in good agreement with experimental values at cell potentials of 2–20 V (corresponding to a current density range of approximately 50–800 A/m²), with mean relative deviation of 9% for low flow rate, narrow electrode gap, polished electrodes, and 150 mg/L NaCl. Highest deviation occurred with a large electrode gap, unpolished electrodes, and Na₂SO₄ electrolyte, due to parasitic H₂O oxidation and less than unity current efficiency. This is the first general model which can be applied to any parallel plate EC system for accurate electrochemical voltage or current prediction.

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1. Introduction

Recent interest has grown significantly in the field of electrochemical coagulation, or electrocoagulation (EC), for drinking or waste waters as an alternative to chemical coagulation (CC) using iron or aluminum salts for a wide range of pollutants, including: organics [1], fluoride [2], arsenic [3,4], and NOM [5–8]. EC has several significant advantages over typical metal salt coagulants (aluminum or ferric chloride/sulfate), including: ubiquity of reactants (industrial iron or aluminum), no chemical addition, no moving parts, and no pH control requirements due to *in-situ* stoichiometric alkalinity generation. A major disadvantage of EC is the required input of electricity. This can make the process uneconomical for treating both drinking water and wastewaters, due to total dissolved solids (TDS) limitations, preventing significant additions of salts (usually NaCl or Na₂SO₄), the most

simple method of increasing solution conductivity (κ) to decrease electrical power consumption. Furthermore, the interdependent relationship between the electrochemical operating parameters and pollutant removal has led to difficulty in the validation of the effectiveness of the EC process.

EC has generally been studied as a pollutant-centric technology, without considerable attention to the underlying electrochemistry [9]. This has likely been due to the electrochemical complexity of EC, involving several inter-related phenomena, including: the cell potential (E_{cell})-current density (i) relationship, the effect of the E_{cell} on EC products, electrokinetic effects, passivation of electrode surfaces, the effect of the electric field ($\nabla\Phi$) on electric double-layer compression in the coagulation process, and the effect of reactor inter-electrode distance (d) and solution flow rate (\dot{V}) on local ion concentrations. The “trial-and-error” engineering is apparent in the wide range of electrochemical operating parameters (E_{cell} , i , and \dot{V}) reported in continuous EC systems (Table 1), previous studies have shown E_{cell} ranges of 2–300 V and i ranges of 2.5–140 A/m². All studies in Table 1 used parallel plate reactor configurations, as more complex reactor designs have shown no significant

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Appendix A. Nomenclature

A	nominal electrode area [m ²]
b_a	anodic Tafel slope [mV dec ⁻¹]
b_c	cathodic Tafel slope [mV dec ⁻¹]
C_p	heat capacity [J mol ⁻¹ K ⁻¹]
CL	charge loading [C L ⁻¹]
CLR	charge loading rate [C L ⁻¹ min ⁻¹]
d	inter-electrode distance [m]
dA	segment electrode area [m ²]
dV	volume between electrodes [m ³]
dy	height of each segment [m]
E^0	equilibrium potential [V]
E_a^0	anodic equilibrium potential [V]
E_c^0	cathodic equilibrium potential [V]
E_a^T	anodic equilibrium potential at temperature T [V]
E_c^T	cathodic equilibrium potential at temperature T [V]
$E_{a,j}$	anodic potential at segment j [V]
$E_{c,j}$	cathodic potential at segment j [V]
E_{cell}	cell potential [V]
E_j	cell potential at segment j [V]
F	Faraday's constant [A·s mol ⁻¹]
Fe_{ris}	residual iron [mg L ⁻¹]
$\dot{H}_{in,j}$	enthalpy flow at inlet of dV at segment j , relative to elements at standard state and 298K [W]
$\dot{H}_{out,j}$	enthalpy flow at outlet of dV at segment j , relative to elements at standard state and 298K [W]
$\dot{H}_{2,j}$	hydrogen flow rate at segment j [m ³ s ⁻¹]
h_f	enthalpy of formation [J mol ⁻¹]
h	height [m]
i	current density [A m ⁻²]
$i_{0,a}$	anodic exchange current density at segment j [A m ⁻²]
$i_{0,c}$	cathodic exchange current density at segment j [A m ⁻²]
$i_{a,j}$	anodic current density at segment j [A m ⁻²]
$i_{c,j}$	cathodic current density at segment j [A m ⁻²]
$I_{Fe,exp}$	experimental current of iron dissolution [A]
$I_{Fe,th}$	theoretical current of iron dissolution [A]
I_j	cell current at segment j [A]
I_{cell}	total cell current [A]
i_j	current density at segment j [A m ⁻²]
i_L	limiting current density [A m ⁻²]
j	segment number [dim]
ML	metal loading [mg L ⁻¹]
n	number of electrode segmentations [dim]
\dot{n}	molar flow [mol s ⁻¹]
$\dot{n}_{H_2,gen,j}$	molar generation rate of hydrogen gas at segment j [mol s ⁻¹]
$\dot{n}_{Fe(OH)_2,gen,j}$	molar generation rate of ferrous hydroxide at segment j [mol s ⁻¹]
$p_{H_2_0}$	initial partial pressure of hydrogen gas [Pa]
p_{H_2n}	reactor exit partial pressure of hydrogen gas [Pa]
pH_0	initial pH [dim]
pH_n	reactor exit pH [dim]
\dot{Q}_j	heat flow entering or exiting dV at segment j [W]
R	Universal gas constant [J mol ⁻¹ K ⁻¹]
R_j	resistance at segment j [Ω]
T	temperature [K]
T_0	initial temperature [K]
T_j	temperature at segment j [K]
T_n	temperature at reactor exit [K]
U_0	superficial velocity of liquid in reactor [m s ⁻¹]

U_n	superficial velocity of liquid at reactor exit [m s ⁻¹]
w	width [m]
\dot{W}_j	work flow entering or exiting dV at segment j [W]
\dot{V}	solution flow rate [m ³ s ⁻¹]
z_a	anode charge transfer number [dim]
z_c	cathode charge transfer number [dim]
α	charge transfer coefficient [dim]
α_k	conductivity temperature coefficient [K ⁻¹]
ζ	mean relative deviation [%]
η	overpotential [V]
η_{Mt}	mass transfer overpotential [V]
$\eta_{a,j}$	anodic overpotential at segment j [V]
$\eta_{c,j}$	cathodic overpotential at segment j [V]
η_k	kinetic overpotential [V]
η_{ohmic}	ohmic overpotential [V]
$\epsilon_{L,0}$	initial liquid volume fraction [dim]
$\epsilon_{L,j}$	liquid volume fraction at segment j [dim]
$\epsilon_{g,j}$	gas volume fraction at segment j [dim]
κ	conductivity [S m ⁻¹]
κ_0	initial conductivity [S m ⁻¹]
κ_n	conductivity at reactor exit [S m ⁻¹]
κ_j	conductivity at segment j [S m ⁻¹]
κ_j^T	conductivity at segment j , adjusted for temperature T [S m ⁻¹]
σ	slip ratio [dim]
ϕ	current efficiency [dim]
$\nabla\Phi$	electric field [V m ⁻¹]
$[Fe^{2+}]_0$	initial ferrous iron concentration [mol m ⁻³]
$[Fe^{2+}]_n$	exit ferrous iron concentration [mol m ⁻³]
ΔS	entropy change [J mol ⁻¹ K ⁻¹]

performance improvement [10]. Modeling of PPERs has included both models of ionic distribution within an EC reactor [11,12] and industrially relevant modeling for specific energy and yield [13]. The E_{cell} - i relationship directly impacts electrical consumption in EC, and has been investigated previously: Vik et al. [5] used a simplified electrochemical potential balance, summing overpotential (η) as a sum of kinetic, mass-transfer, and ohmic resistance terms, but had no experimental validation; Chen et al. [14] formed a complex potential balance that grouped all unknown variables, including Tafel parameters and overpotential terms, into experimentally derived constants – rendering the model inapplicable to other reactor geometries; Mechelhoff [15] constructed a two-dimensional finite element model which simultaneously solved the Navier-Stokes and Nernst-Planck equations for current density distribution, although no experimental validation was attempted; Zongo et al. [16] experimentally fitted the E_{cell} - i relationship to find $E_{cell} = 0.1 + (d/\kappa)i + 0.20 \ln(i)$ for both Al and Fe electrodes in a recirculating continuous system, but again the model is not appropriate for other geometries; Izquierdo et al. [17] experimentally determined a “corrected” equilibrium voltage (E^0) by assuming a constant η_{ohmic} , which only applied to the author's experimental cell. Most of these prior studies ignored the effects of cathodic H₂ generation and temperature rise due to joule heating, and reduced the η terms and Butler-Volmer (BV) equation to experimental constants. This, unfortunately, has prevented these models from being generalized for scale-up or validation in other systems. A general E_{cell} - i relationship, including the impact of cathodic H₂ generation and heat generation, derived from only tabulated constants and arbitrary dimensions in a parallel EC system, would thus be extremely valuable. Our objectives were to derive a general E_{cell} - i relationship, and to test the robustness of the model by

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