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Research article

Numerical simulation and exploration of electrocoagulation process for arsenic and antimony removal: Electric field, flow field, and mass transfer studies

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ABSTRACT

In order to intuitively and clearly evaluate the potential and current distribution, the fluid flow and mixing, as well as mass transfer involved in electrocoagulation process for As and Sb removal, numerical simulation of electric field, flow field and mass transfer were constructed by Comsol Multiphysics and verified by experiments. Results displayed that the primary current and potential distribution were improved by changing electrode distance or adding insulator in a batch reactor. When configuration 2 and 2 cm electrode distance were applied, a more uniform primary current distribution and higher electrode current efficiency were obtained. In a continuous flow reactor, the increase of flow rate resulted in the left shift of the peak in residence time distribution curve, gradual decrease of the tailing area, reduction of the stagnation zone, and more uniform mixing of the fluid. However, higher than 0.043 L/min was unfavorable to the formation of flocs and its effective combination with pollutants. According to the simulation of mass transfer, at the initial stage, the rate of electrolysis/hydrolysis was greater than that of mass transfer. Fe^{2+}, OH^{-}, and $Fe(OH)_2$ were primarily concentrated on the anode, cathode, and between the two electrodes, respectively. Under the action of electromigration, diffusion and convection, the concentration distribution of $Fe(OH)_2$ increased at the direction of streamline. The concentration of Fe^{2+} and OH^{-} achieved the minimum value at the outlet. However, $Fe(OH)^{+}$ concentration and distribution were hardly affected by the treatment time, and once generated, immediately proceed to the next hydrolysis reaction.

1. Introduction

Arsenic (As) and antimony (Sb), toxic and carcinogenic trace elements present in drinking water, poses a great threat to human beings (Wu et al., 2011; Kang et al., 2011). In recent years, electrocoagulation (EC) has been widely applied in wastewater treatment especially in heavy metal removal (Lai and Lin, 2003; Amrose et al., 2014). Most researches focused on the evaluation and optimization of operating conditions, but little attention was paid to the exploration of the EC process and optimization of the EC reactor (Sahu et al., 2014; Song et al., 2017). It contains several physicochemical processes, such as electro-oxidation, electro-reduction, electro-flotation, coagulation or co-precipitation, which play a vital role in treatment efficiency and mechanism study of EC system (Bazrafshan et al., 2015). However, these complex processes are difficult to analyze accurately only by experimental methods, or have long experimental periods and high operating costs. In fact, there are flow field, electric field as well as temperature field in EC process, involving mass transfer, fluid flow, electron transfer and so on (Song et al., 2017; Hakizimana et al., 2017). They are closely related to the physicochemical reactions, and can be mathematically described by partial differential equations (PDEs) (Dickinson et al., 2014; et al., Huang, 2010). Therefore, in order to obtain the detailed information of flow field distribution, potential and current distribution as well as mass transfer during EC process, the need for exploration of EC reactor configuration and reaction process from a more microscopic view is very essential.

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Based on our previous studies about As and Sb removal by EC process (Song et al., 2015, 2017; Huang, 2010), in this paper, COMSOL Multiphysics, on the basis of the finite element method (Dickinson et al., 2014; CnTech, 2007; Wang and An, 2012), was employed to solve PDEs for multiphysics simulation to visually discuss the electric field distribution, flow field distribution and mass transfer of EC process for As and Sb removal.

According to the mass conservation equation of electrolyte and current conservation equation, the electrochemical module of COMSOL was applied to simulate and evaluate the potential and current distribution in EC process. Consequently, the configuration of batch EC reactor was improved and a more uniform primary current distribution was obtained, which improved the EC performance for As and Sb removal. On the basis of the mass conservation equation, momentum conservation equation and energy conservation equation, the CFD module of COMSOL was used to simulate the velocity field and residence time distribution in the flow field of EC process, and the continuous flow EC reactor was optimized. In the light of the Navier-Stokes (N-S) equation and Nernst-Planck (N-P) equation, the chemical transfer model of COMSOL was employed to explore the mass transfer, diffusion and migration process. Besides, experimental verifications were also performed. As a result, the potential and current distribution, fluid mixing in EC reactor, and concentration and distribution of EC products in EC process were intuitively evaluated, which optimized the EC reactor configuration, saved the research time and reduced the operating costs to the greatest extent.

In conclusion, Numerical simulation by COMSOL coupling with experimental verification of EC process not only developed a new way for optimization and exploration of the EC process, but also deepened the understanding of EC performance and further improved the theoretical basis of EC system.

2. Formulation of numerical simulation

In this study, COMSOL Multiphysics 5.2, the finite element commercial software, is employed to solve the nonlinear PDEs (Dickinson et al., 2014; CnTech, 2007; Wang and An, 2012) and construct the numerical simulation of electric field, flow field and mass transfer of the EC system. The model converges when the weighted absolute residual norm is less than 10^{-6} (COMSOL AB, 2016).

2.1. Potential and current distribution

In the simulation of EC electric field, the physical equations involve in the mass conservation equation and current conservation equation as follows (Wu, 2008; Vázquez et al., 2012; Dubrawski et al., 2014):

Mass conservation equation in electrolyte solution:

$$\frac{\partial c_i}{\partial t} + \nabla \cdot N_i = R_{i,\ tot} \tag{1}$$

$$N_i = -D_i \nabla c_i - z_i u_{m,i} F c_i \nabla \varnothing_l + c_i u \tag{2}$$

Among them, $D_i \nabla c_i$ is the diffusion term, $z_i u_{m,i} F c_i \nabla \varnothing_l$ is the electromigration term, $c_i u$ is the convection term, and $R_{i,\ tot}$ is the term of additional reaction source.

The amount of substance is replaced by the quantity of electricity:

$$i_l = F \sum_{i=1}^{n} z_i (-D_i \nabla c_i - z_i u_{m,i} F c_i \nabla \varnothing_l) \tag{3}$$

Besides, electric neutrality satisfies the condition of $\sum z_i c_i = 0$.

Current conservation equation in electrolyte solution:

$$\nabla \cdot i_l = Q_l + F \sum_i z_i R_i \tag{4}$$

Q_l is the additional current source.

The properties and boundary conditions in this model were displayed in Table 1. The diagram of three dimensional configuration and cross sections of batch EC reactor of this model were shown in Figs. S1(a and b).

Table 1
The properties and boundary conditions involved in the model.

Name	Equation
Electrolyte	$\nabla \cdot i_l = Q_l$ $i_l = -\sigma_l \nabla \varnothing_l$
Electrode	$\nabla \cdot i_s = Q_s$ $i_s = -\sigma_s \nabla$
Electrode-Electrolyte Boundary Interface	$\varnothing_{s,ext} - \varnothing_l = E_{eq}$
Insulation	$-n \cdot i_l = 0$ $-n \cdot i_s = 0$
Boundary	$\varnothing_{s,ext} = 0$

l and s refers to the electrolyte and electrode, respectively; $\varnothing$(V) refers to the potential; σ(S/m) refers to the conductivity; and E_{eq}(V) refers to the equilibrium potential.

2.2. Hydrodynamics analysis

Within the scope of Newtonian fluid, the flow phenomena can be described by the following equations (Vázquez et al., 2010, 2010, 2014; Vázquez et al., 2010):

Mass conservation: $$\frac{\partial \rho}{\partial t} + \nabla(\rho \cdot u) = 0 \tag{5}$$

Momentum conservation: $$\rho \frac{\partial u}{\partial t} + \rho(u \cdot \nabla)u = \nabla \cdot [-p \cdot I + \tau] + F \tag{6}$$

Energy conservation:

$$\rho \cdot C_p \left(\frac{\partial T}{\partial t} + (u \cdot \nabla) T \right) = -(\nabla \cdot q) + \tau : S - \frac{T}{\rho} \frac{\partial p}{\partial T} \Big|_p \left(\frac{\partial \rho}{\partial t} + (u \cdot \nabla) p \right) + Q \tag{7}$$

Among them, ρ is the fluid density, t is the time, u is the flow velocity, p is the pressure, F is the volume force, C_p is the constant pressure specific heat capacity, T is the absolute temperature, q is the heat flux, τ is the viscous shear stress tensor, and $\tau = \begin{pmatrix} \tau_{xx} & \tau_{xy} & \tau_{xz} \\ \tau_{yx} & \tau_{yy} & \tau_{yz} \\ \tau_{zx} & \tau_{zy} & \tau_{zz} \end{pmatrix}$, : refers to the operator, and a: b= $\sum_n \sum_m a_{nm} b_{nm}$, S is the strain rate tensor, and $S = \frac{1}{2}(\nabla u + (\nabla u)^T)$, and Q is the heat source.

The continuous flow reactor with 2 cm electrode distance was simulated, and the three dimensional configuration and cross sections of continuous EC reactor were shown in Figs. S1(c and d). Reynolds number was less than 2300, and therefore the laminar flow interface of CFD module was employed in this model.

The fluid properties in this model should follow Eqs. (8) and (9), and F (N/m^3) refers to the volume force,

$$\rho(u \nabla)u = \nabla \cdot [-pI + \mu(\nabla u + (\nabla u)^T)] + F \tag{8}$$

$$\rho \nabla \cdot (u) = 0 \tag{9}$$

The wall of EC reactor satisfied the condition of $u = 0$, and the boundary condition was no slip. Settings of inlet, outlet and symmetry followed Eqs. (10)–(12), respectively. The pressure of outlet was 0 Pa, and suppress backflow was set.

$$u = -u_0 n \tag{10}$$

$$[-pI + \mu(\nabla u + (\nabla u)^T)]n = -\widehat{p_0} n \widehat{p_0} \leq p_0 \tag{11}$$

$$u \cdot n = 0,\ \kappa - (\kappa \cdot n)n = 0,\ \kappa = [\mu(\nabla u + (\nabla u)^T)]n \tag{12}$$

Among them, κ is the turbulent kinetic energy, μ is the dynamic viscosity, and n is the kinematic viscosity.

After setting the above properties and boundary conditions, physics-