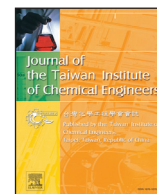




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Construction and optimization of an iron particle–zeolite packing electrochemical–adsorption system for the simultaneous removal of nitrate and by-products

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ARTICLE INFO

Article history:

Received 3 December 2017

Revised 10 February 2018

Accepted 14 February 2018

Available online xxx

Keywords:

Electrochemical–adsorption (ECA) system

Nitrate and by-products

Optimization

Polarized iron particle

Zeolite

ABSTRACT

In this study, an electrochemical–adsorption (ECA) system was constructed by packing iron particles and zeolites between electrodes to simultaneously remove nitrate and its by-products without excessive active chloride accumulation. Response surface methodology (RSM) with Box–Behnken design (BBD) was applied to investigate the effects of independent variables (iron particle, zeolite and current density) and their interaction on the system performance, also determining its optimum working state. The optimal conditions for the amount of iron particles (19.74 g) and zeolites (28.19 g) as well as the current density (18.72 mA/cm²) resulted in a high nitrate removal efficiency of 95% and merely a little ammonia accumulation. Polarized iron particles could provide more reactive sites and increase mass transfer efficiency, thus promoted nitrate reduction and decreased energy consumption. Synergistic effects of electrolysis and zeolite adsorption accelerated ammonium removal by increasing the rate of ion directional migration. The system having low operation costs and no secondary pollution appeared to be an advisable enhancing strategy for removing nitrate and by-products.

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1. Introduction

Nitrate, a common contaminant, is frequently detected in water bodies and nitrate contamination of groundwater has become a widespread environmental issue [1]. High nitrate concentrations in drinking water can result in serious health issues [2]. Currently, a number of technologies including ion exchange [3], heterogeneous catalysis [4], biological denitrification [5], and electrochemical process [6] have been developed to reduce nitrate concentration in groundwater below the maximum limits (10 mg-N/L NO₃⁻) stipulated by the World Health Organization (WHO) [7]. Among these methods, electrochemical processes are particularly suitable for nitrate removal owing to their high treatment efficiency, easy operation, and environmental friendliness [8]. However, traditional electrochemical systems comprised of cathode and anode typically convert 60–70% of nitrate by cathodic reduction [6]. With the aim to enhance the electrochemical nitrate reduction efficiency, iron particles and iron nets have been used between the electrodes

serving as bipolar electrodes for promoting nitrate reduction [9,10]. With regard to the main by-products of nitrite and ammonia, these compounds were increasingly produced as the efficiency of the nitrate transformation process increased. Having this in mind, different anode materials were used to remove nitrite and ammonia *via* oxidation by hydroxyl radicals [32]. However, hydroxyl radicals are unstable, which was not beneficial to nitrite and ammonia oxidation during the entire electrochemical process. Hence, NaCl has been widely utilized in electrochemical systems to remove nitrite and ammonia *via* generation of active chloride that had the excellent oxidizability for oxidizing nitrite and ammonia [6]. However, the formation of active chloride in excess can contaminate the electrolyte. Meanwhile, the concentration of NaCl in polluted water has been demonstrated to decrease in inland regions [11]. Therefore, the addition of NaCl for the removal of by-products is an unsustainable solution with respect to the operating costs [12].

Zeolites are known to have excellent cation exchange capacities, especially for ammonia. As previously demonstrated [14], zeolites present optimum performance toward the adsorption of ammonia at different experimental conditions than composite and active carbon. Thus, this kind of material has been widely used to remove ammonia from wastewater by adsorption processes [13].

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<https://doi.org/10.1016/j.jtice.2018.02.023>

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Table 1
Experimental design and response matrixes.

Run	Factor A: iron particle		Factor B: zeolite		Factor C: current density		Experimental results		
	Code	Actual (g)	Code	Actual (g)	Code	Actual (mA/cm ²)	NO ₃ ⁻ (mg/L)	NO ₂ ⁻ (mg/L)	NH ₄ ⁺ (mg/L)
1	0	12.5	0	15	0	12.5	2.01 ± 0.27	0.08 ± 0.01	16.94 ± 1.88
2	-1	0	1	30	0	12.5	10.17 ± 1.13	0.52 ± 0.08	7.51 ± 1.53
3	0	12.5	0	15	0	12.5	2.25 ± 0.23	0.07 ± 0.01	15.87 ± 2.56
4	0	12.5	-1	0	1	20	0.93 ± 0.12	0.08 ± 0.01	12.61 ± 1.58
5	0	12.5	1	30	-1	5	8.41 ± 0.58	0.35 ± 0.05	20.10 ± 1.16
6	0	12.5	0	15	0	12.5	1.93 ± 0.28	0.08 ± 0.01	16.89 ± 3.67
7	-1	0	-1	0	0	12.5	6.81 ± 0.46	0.32 ± 0.06	24.34 ± 2.72
8	-1	0	0	15	1	20	10.83 ± 1.31	0.16 ± 0.02	5.21 ± 0.84
9	0	12.5	0	15	0	12.5	2.10 ± 0.42	0.07 ± 0.01	16.27 ± 1.13
10	1	25	0	15	-1	5	8.40 ± 0.41	0.27 ± 0.07	27.90 ± 0.60
11	1	25	0	15	1	20	2.23 ± 0.33	0.06 ± 0.01	8.27 ± 0.67
12	-1	0	0	15	-1	5	16.75 ± 1.47	0.57 ± 0.09	20.77 ± 1.41
13	0	12.5	0	15	0	12.5	2.18 ± 0.20	0.07 ± 0.01	16.50 ± 0.64
14	1	25	-1	0	0	12.5	1.99 ± 0.39	0.16 ± 0.08	21.79 ± 1.87
15	0	12.5	1	30	1	20	5.32 ± 0.64	0.01 ± 0.01	3.43 ± 0.61
16	0	12.5	-1	0	-1	5	8.72 ± 0.96	0.44 ± 0.01	31.98 ± 3.75
17	1	25	1	30	0	12.5	2.89 ± 0.59	0.11 ± 0.05	9.59 ± 1.31

However, zeolites have been reported to require long periods (e.g., 7 h) for reaching the adsorption equilibrium as a result of the relatively low ammonia removal rates [15]. Ding et al. added zeolite to an electrolytic reactor to construct a zeolite-packed electrochemical ammonia removal system. About 69% of the initial ammonia (20 mg-N/L) was removed via electrochemical oxidation and zeolite adsorption within 1 h [16]. An electrochemical ion-exchange reactor containing zeolites achieved efficient ammonia removal and zeolite regeneration in the presence of NaCl during electrolysis [33]. As revealed by Huang et al. [31], zeolites maintained superior ammonia adsorption capacities during electrolysis because of the high stability of the zeolitic material [31]. However, the high energy consumption, resulting from the poor electrical conductivity of zeolites, hindered the applications of this zeolite-packed electrochemical system. On the other hand, although numerous researchers have attempted to remove ammonia by combining electrolysis and adsorption processes, this method has not been used for the simultaneous removal of nitrate and the ammonia by-product. The previous study found that iron particles placed between the electrodes could be polarized for decreasing energy consumption, also providing more reactive sites for promoting nitrate reduction [9]. Therefore, utilization of an electrochemical system with iron particles and zeolites can be a feasible approach for efficiently removing nitrate and its by-products of ammonia, nitrite with relatively low energy consumption.

Several parameters have been identified to determine the electrochemical removal efficiencies of nitrate and its by-products to a large extent, and attempts to optimize the electrolytic process have been made. In this sense, response surface methodology (RSM) is an efficient statistical technique for optimizing multiple variables with a small number of experiments [17]. Box-Behnken design (BBD) was proved to be suitable for identifying correlations between various variables and determining favorable conditions [19]. While, RSM and BBD have been successfully utilized in a wide range of areas [20], the combination of both methods has not been carried out yet for optimizing the operational parameters in intensified electrochemical-adsorption (ECA) systems.

In this study, an ECA system was first fabricated by packing iron particles and zeolites between the electrodes. This system allowed us to simultaneously remove nitrate and its by-products of ammonia and nitrite, as well as reducing energy consumption without excessive active chloride accumulation. RSM and BBD methods were applied to predict the nitrate, nitrite, and ammonia removal capacities of the as-built ECA system under varying

operating conditions (e.g., amount of iron particles, zeolite, and current density) and to determine its optimum working state. Meanwhile, the mechanisms behind the removal of nitrate and its by-products in this system were also investigated.

2. Experimental

2.1. Synthetic contaminated groundwater

Synthetic nitrate solutions were prepared from a 0.303 g/L NaNO₃ to a final concentration of 50 mg-N/L, according to the mean nitrate content of groundwater in rural areas of Dalian [24]. 0.5 g/L Na₂SO₄ was added into the electrolytes to increase the solution conductivity. The chemical reagents used in the experiments were of analytical grade (Sinopharm, China) and the solutions were prepared with deionized water (18.2 MΩ/cm).

2.2. Experimental materials and apparatus

Metallic iron spherical particles of 3 mm in diameter were purchased from a local factory (Beijing, China). Zeolites (8–10 mesh, 1.5–2.5 mm) were obtained from Wako Pure Chemical Industries. Before electrolysis, zeolites were washed with deionized water to remove ultrafine particles and subsequently oven dried at 60 °C.

A 500 mL (4.5 cm × 7.0 cm × 15.0 cm) rectangular undivided cell was manufactured with an acrylic material. Water samples were obtained from a sampling valve located at the middle of the reactor. Cu–Zn (Cu: 60 wt%; Zn: 40 wt%) cathode and Ti/IrO₂–Pt anode (IrO₂: 78 wt%; Pt: 22 wt%) (TohoTech company, Japan) of 64.5 cm² (15 cm × 4.3 cm) were utilized in all the processes separated with a distance of 4 cm. Iron particles and zeolites were uniformly mixed and loaded in a perforated container (9.0 cm × 5.0 cm × 3.0 cm, punched holes of 2 mm in diameter on four of the sides, hole distribution of 6/cm²) placed between the electrodes to construct the ECA system. A DC power (Takasago, EX1500H2) with a voltage range of 0–50 V and a current range of 0–5 A was employed for providing constant current.

2.3. Experimental procedure

2.3.1. Batch electrolysis for determining optimum operating conditions

As shown in Table 1, several experiments were performed in the ECA system. 250 mL of the synthetic nitrate solution was

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