Journal of Water Process Engineering 9 (2016) 254–266

Contents lists available at ScienceDirect

Journal of Water Process Engineering

journal homepage: www.elsevier.com/locate/jwpe

Nitrate removal using poly-*o*-toluidine zirconium(IV) ethylenediamine as adsorbent: Batch and fixed-bed column adsorption modelling

Nafisur Rahman*, Mohammad Fazeel Khan

Department of Chemistry, Aligarh Muslim University, Aligarh, Uttar Pradesh 202002, India

ARTICLE INFO

Article history:
Received 13 August 2015
Received in revised form 9 January 2016
Accepted 10 January 2016
Available online 24 January 2016

Keywords:
Poly-*o*-toluidine zirconium(IV) ethylenediamine
Nitrate
Fixed-bed column
Breakthrough curves
Desorption

ABSTRACT

Nitrate removal from aqueous solutions was studied using poly-*o*-toluidine zirconium(IV) ethylenediamine (PTZE) in batch and fixed bed column experiments. The kinetic data obtained from batch experiments were analyzed by pseudo first order, pseudo second order, Elovich, intraparticle diffusion, Boyd's and Bangham's models. The adsorption data were well described by pseudo second order model. The effects of operating parameters such as flow rate (1–3 mL min^{-1}), bed height (1–3 cm) and concentration (100–500 mg L^{-1}) were studied in column mode. The adsorption efficiency increased with increasing initial concentration and bed height and decreased with increasing flow rate. The obtained experimental data were in good agreement with Thomas and Yoon–Nelson models, but in the case of Bohart–Adams model, low correlation coefficient was observed. Desorption of nitrate from PTZE loaded nitrate was carried out with 0.15 M NaOH and regenerated with 1 M HCl solution. The adsorbent can be used successfully upto 8 cycles of sorption–desorption processes.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Nitrate is the most widespread contaminant of ground and surface waters all over the world. Agricultural over-application of natural and synthetic fertilizers, aquaculture, municipal wastewater, human and animal waste, overflowing septic tanks, processed food, dairy and meat products, detergent manufacturing and mineral processing industries are the sources of nitrate into the aquatic environment [1]. High level of nitrate in drinking water may cause adverse health effects such as blue baby syndrome [2,3], especially in infants, cyanosis and cancer of the alimentary canal [4], and diabetes [5]. The European Community established a maximum contaminant level of 50 mg/L and recommended level of 25 mg/L [6]. Therefore, the monitoring and removal of nitrate from water are relevant due to its potential dangerous impact on both environment and human health.

The conventional methods used for the removal of nitrate can be divided into three main groups: physical, chemical and biological treatment processes [7–11]. Among them, adsorption methods are generally considered to be more promising and effective because they allow simple and economical operation, resulting in less sludge production and disposal problems. In recent years, several studies have been performed to explore the application of zirconium(IV) based materials for adsorption of anionic pollutants [12,13]. In our earlier studies [14], PTZE was synthesized which showed affinity for nitrate in the pH range 3–6. The equilibrium sorption data fitted well to Freundlich isotherm model. A single-stage batch adsorption system was developed from Freundlich adsorption isotherm parameters for removal of nitrate from water. A new inorganic-sugar beet pulp composite was prepared from sugar beet pulp after loading with zirconium(IV) ions and used for removal of nitrate from water [15]. The adsorption of nitrate by chitosan hydrobeads was increased with increase in the pH of solution and depends on the temperature with an optimum activity at 30 °C. The kinetic results corresponded well with the pseudo second order rate equation and the intraparticle diffusion also played a significant role in the initial stage of adsorption process [16].

Arora et al. [17] have modified the surface of natural zeolite by coating with a chitosan layer. The chitosan coated zeolite was protonated and used as adsorbent for removal of nitrate from water. Nitrate can also be removed from aqueous solution by modified clinoptilolite zeolite. The investigation of kinetic equations indicated that nitrate adsorption followed pseudo first order kinetic model [18]. Adsorption of nitrate from aqueous solutions on ammonium functionalized mesoporus MCM-48 silica was investigated [19,20]. The adsorbent was prepared via a post synthesis grafting method using aminopropyltriethoxysilane. At ambient temperature, the removal of nitrate was maximum at pH 8.

* Corresponding author.
E-mail address: nafisurrahman05@gmail.com (N. Rahman).

http://dx.doi.org/10.1016/j.jwpe.2016.01.007
2214-7144/© 2016 Elsevier Ltd. All rights reserved.

Zn/Al chloride layered double hydroxide was prepared and used as sorbent for removal of nitrate [21]. Recently, the combination of magnetic Υ–Fe_2O_3 nanoparticles with a coating of hydroxyapatite has been applied for the removal of nitrate from environmental water and soil samples [22]. Under the optimum conditions, the adsorption efficiency of the adsorbent was in the range of 93–101%. Hydrous bismuth oxide has been used for the removal of nitrate [23]. The adsorption of nitrate onto HBO_2 followed pseudo first order kinetic model more closely compared with pseudo second order kinetic model. As the temperature of the solution increased, the adsorption capacity of the HBO_2 also increased, indicating the endothermic nature of adsorption process. Adsorption feasibility of meso alumina and nano alumina has been assessed for nitrate removal from industrial effluent samples. Rate kinetics of adsorption phenomenon depicts pseudo second order kinetic model. Intraparticle diffusion and Elovich kinetic fail to explicit effectively the adsorption data [24]. Ahmad et al. [25] have synthesized alumina supported nano zero valent zinc and used as adsorbent for removal of arsenic and nitrate from water. The adsorption data fitted well to pseudo second order kinetic model. Ji et al. [26] have investigated the feasibility of hybrid system of zero valent iron combined with adsorbents for the removal of nitrate from water. The maximum nitrate removal in combined Fe°-granular activated carbon, Fe°-filtralite and Fe°-sepiolite systems was 86, 96 and 99%, respectively.

Iron impregnated acrylamide matrix was used to treat the pharmaceutical wastewater in a fluidized bed reactor as heterogeneous catalyst [27]. The biomaterials have also been employed for removal of pollutants from wastewater. An indigenous mixed bacterial culture was used to decolorize acid red 88 dye under static condition. The decolorization parameters were optimized using Taguchi's OA experiments [28]. Base activated Setaria verticillata carbon was found to be a promising adsorbent for the removal of red yellow 15 dye from aqueous solution [29]. Most studies on nitrate removal by adsorbents have been carried out in batch mode. However, few studies have been reported for removal of nitrate in fixed bed columns [30–32]. The advantages associated with fixed bed column experiments include (i) the direct application to get reliable solution for designing optimization and (ii) providing breakthrough curves of fixed bed columns in the removal of nitrate from real water treatment processes.

The purpose of this investigation was to gain an insight into the removal efficiency of nitrate by PTZE in both batch and fixed-bed systems. The objectives of this study were divided into the following parts:

(i) Analysis of experimental data from batch adsorption by pseudo-first order, pseudo-second order, Elovich, intraparticle diffusion, Boyd's and Bangham's adsorption kinetic equations.
(ii) Investigation of effects of column operating parameters such as flow rate, bed height and initial concentration on the breakthrough curves.
(iii) Analysis of experimental data from column adsorption using Bohart–Adams, Thomas and Yoon–Nelson models, and
(iv) Desorption of nitrate from PTZE loaded nitrate to evaluate the possibility of reuse.

2. Experimental

2.1. Reagents and instrument

Zirconium oxychloride octahydrate (Otto Chemie. Pvt., Ltd., India), ethylenediamine (Merck, India), *o*-toluidine (Sd. Fine Chem. Ltd., India) and ammonium persulphate (Thomas Baker Chemicals Pvt., Ltd., India) were used for the synthesis of poly-*o*-toluidine zirconium(IV) ethylenediamine. All other chemicals and reagents used were of analytical reagent grade. Double distilled water was used for all the studies. A digital ion-analyzer (Elico LI 126 India) and Eutech digital pH meter (Cyberscan pH 2100) were used for measuring the concentration of nitrate ion and pH of the solution, respectively.

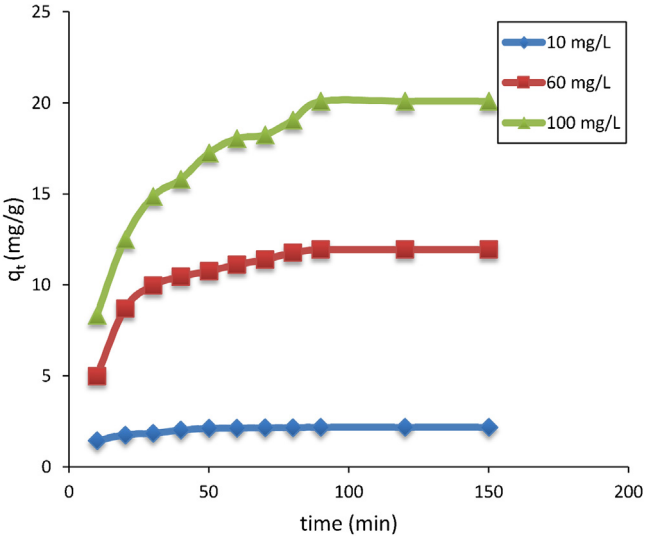

Fig. 1. Effect of contact time on adsorption capacity of nitrate (adsorbent dose = 0.2 g, temperature = 27 °C, pH 4.5).

2.2. Preparation of poly-o-toluidine zirconium(IV) ethylenediamine (PTZE)

PTZE was synthesized according to previously described procedure [14]. A white gelatinous precipitate of zirconium (IV) ethylenediamine was prepared by adding equal volumes of 0.1 M zirconium oxychloride solution and 0.1 M aqueous ethylenediamine at 27 °C. Poly-*o*-toluidine was prepared by mixing equal volumes of 0.4 M ammonium persulphate in 4 M HCl and 20% *o*-toluidine prepared in 2 M HCl with constant stirring on a magnetic stirrer at 0 °C. A green colored gel was obtained. Further, the gel of poly-*o*-toluidine was added to the white gelatinous precipitate of zirconium(IV) ethylenediamine and pH of the resulting mixture was adjusted to 2.0. It was stirred on a magnetic stirrer for 10 h and left at room temperature for another 24 h for digestion. The PTZE gel was filtered off, washed thoroughly with distilled water to remove excess acid and any other adhering trace of ammonium persulphate. The washed gel was dried in an oven at 50 °C and then the product was immersed in distilled water to obtain small granules. The material was converted into form by treating with 1 M HCl solution for 24 h. The excess was removed by several washings with distilled water. The material was finally dried in an oven at 50 °C and sieved to obtain particles of uniform size range (~125 μm).

2.3. Batch adsorption experiments

The adsorption experiments were performed by batch process. Fixed amount of PTZE (0.2 g) was added in the stoppered conical flasks containing 50 mL of nitrate ion solution of different concentrations (10, 60 and 100 mgL^{-1}) at pH 4.5. The mixture was shaken for the desired contact time in an electrically thermostated reciprocating shaker at the rate of 120 strokes/min at 27 ± 1 °C. At the regular interval of time, the conical flask from the shaker was withdrawn and then the adsorbent was separated from the nitrate ion

Download English Version:

https://daneshyari.com/en/article/232488

Download Persian Version:

https://daneshyari.com/article/232488

Daneshyari.com