

Spring 5-2-2011

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Recommended Citation

Ammar Arab Beddai and Basava Rao, "Removal of Nitrate from Water Using Fluidized Bed Ion Exchange Column" in "10th International Conference on Circulating Fluidized Beds and Fluidization Technology - CFB-10", T. Knowlton, PSRI Eds, ECI Symposium Series, (2013). <http://dc.engconfintl.org/cfb10/18>

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REMOVAL OF NITRATE FROM WATER USING A FLUIDIZED BED ION EXCHANGE COLUMN

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ABSTRACT

Experimental and theoretical studies were carried out to investigate the performance of a fluidized bed ion exchange system to remove nitrate. The exchange of nitrate on strong anion exchange resin (TULSION A-27) was studied in the flow rate range of 2 to 7 L/h. Nitrate removal was done at five conditions of the expanded bed height (Z). The results showed that the experimental data can be fitted to Richardson and Zaki equation, and the comparison between the experimental and calculated terminal velocities showed low relative error.

INTRODUCTION

Increased levels of nitrate in ground water have made many wells unsuitable as sources for drinking water. Nitrate is so toxic, especially to pregnant women and infants, that the USEPA (United States Environmental Protection Agency) standard of 10 mg NO₃⁻N/L or less in drinking water was established for human health [Taekyung *et al* (1); Kavita *et al* (2); Lucija *et al* (3)]. Ion exchange is a chemical treatment process used to remove unwanted ionic species from wastewater [M. Matosic *et al* (4); Robert Kunin (5); Ammar Arab (6)].

Solid-liquid fluidized beds (SLFB) are used in industry for hydrometallurgical operations, catalytic cracking, chromatographic separation, ion exchange, adsorption, crystallization and sedimentation, etc [Prakash and Jyeshtharaj (7); Srikuma *et al* (8)]. However, the packed bed ion exchange process has some disadvantages such as high pressure drop and bed clogging. These disadvantages can be eliminated if the packed bed is replaced by a fluidized bed [Shyh-Jye and Wen-Jang (9); Hideaki *et al* (10); Seung-Jai *et al* (11)]. The purpose of this study was to investigate nitrate removal in a liquid-solid fluidized bed ion-exchange system. The effects of operating parameters including liquid flow rate and height of the bed on the removal rate of nitrate were studied. The experimental data of voidage versus superficial velocity were successfully correlated using the Richardson-Zaki Equation.

THEORY OF FLUIDIZED BED

Minimum fluidization velocity

The minimum fluidization velocity can be obtained using the Ergun Equation by setting the pressure drop equal to the pressure required to suspend the bed or the weight of the bed divided by the cross sectional area [McCabe *et al* (12); Vassilis *et al* (13)]:

$$\frac{150\mu u_{fm}}{\Phi_s^2 d_p^2} \frac{1-\varepsilon_{fm}}{\varepsilon_{fm}^3} + \frac{1.75\rho u_{fm}^2}{\Phi_s d_p} \frac{1}{\varepsilon_{fm}^3} = g(\rho_p - \rho) \quad (1) \quad \text{and} \quad \text{Re}_{fm} = \frac{u_{fm} \rho d_p}{\mu} \quad (2)$$

Estimation of bed voidage (ε)

Bed voidage was obtained by substitution of data into the following equation [Mohsen *et al* (14)]:

$$\varepsilon = \left[1 - \frac{V_p}{V_L} \right] = \left[1 - \frac{V_p}{A_T \cdot H} \right] = \left[1 - \frac{m_p}{\rho_p \cdot A_T \cdot H} \right] \quad (3)$$

Bed expansion characteristics

Before processing with feedstock, determination of the bed expansion characteristics was necessary. It was decided to use the well-known Richardson–Zaki Equation [Mohsen *et al* (14)]:

$$\frac{U}{U_t} = \varepsilon^n \quad (4)$$

The expanded bed height is [Inglezakis and Pouloupoulos (15)]: $Z_f = Z \frac{1-\varepsilon}{1-\varepsilon_f}$ (5)

The average bed voidage can be estimated from Eq. (3) or the following equation:

$$\varepsilon_f = 1 - (1-\varepsilon) \frac{Z}{Z_f} \quad (6)$$

Where $\varepsilon_0=0.41$ is the packed-bed voidage. Values of the Richardson–Zaki coefficients n , and the apparent terminal velocities of the particles U_t were calculated from linear regression of plots of $\ln(U)$ versus $\ln(\varepsilon)$. For the particles which had a mean diameter of 750 μm , the apparent terminal velocities of the particles U_t were calculated from Allen's equation, Eq. (7). For adsorbents with an approximate mean particle size of 200 μm one can calculate U_t using Stokes' law. U_t Stokes is given by Eq. (8) [Mohsen *et al* (14)].

$$U_t = 0.27 \sqrt{\frac{d_p (\rho_p - \rho) g}{\rho}} \text{Re}_t^{0.4} \quad (7)$$

$$U_{t-stokes} = \frac{g (\rho_p - \rho) d_p^2}{18\mu} \quad (8)$$

Where (n) is the index which depends on the particle size and sedimentation velocity. The expansion index (n) was calculated by following equations [Yong-Hong *et al* (16)]:

$$\begin{aligned} n &= 4.65 & \text{Re}_i < 0.2 \\ n &= 4.4 \text{Re}_i^{-0.03} & 0.2 < \text{Re}_i < 1 \\ n &= 4.4 \text{Re}_i^{-0.1} & 1 < \text{Re}_i < 500 \\ n &= 2.4 & 500 < \text{Re}_i \end{aligned} \quad (9)$$

The expansion behavior can also be estimated from empirical considerations. Hartmann obtained a good fit of their experimental data for n using Eq. (10) [Mohsen *et al* (14)].

$$n = \frac{5.09 + 0.2309 \text{Re}_i^{0.877}}{1 + 0.104 \text{Re}_i^{0.877}} \quad (10)$$

The model of Shiller and Naumann is commonly used for the prediction of terminal velocity of a spherical particle [M.H.Shahavi *et al* (17)]:

$$G_a = 18 \text{Re}_i + 2.7 \text{Re}_i^{1.687} \quad 3.6 < G_a < 10^5 \quad (11)$$

Where the Grashof number is given by the following equation:

$$G_a = \frac{\rho (\rho_p - \rho) g d_p^3}{\mu^2} \quad (12)$$

The fluidization behavior of particles is based on the Gallileo number [Vassilis *et al* (13)]:

$$Ga = \frac{\rho_p g (\rho_p - \rho) d_p^3}{\mu^2} \quad (13)$$

From Ga the terminal velocity Reynolds number can be calculated for different column and particle diameters:

$$\text{Re}_i = \left[\frac{23}{Ga} + \frac{0.6}{Ga^{0.5}} \right]^{-1} \left[1 + 2.35 \frac{d_p}{d_c} \right]^{-1} \quad (14)$$

The expansion index was estimated from Ga:

$$\frac{5.1 - n}{n - 2.4} = 0.016 Ga^{0.67} \quad (15)$$

For the relatively large particles (of several millimeters) in water, the equation proposed by Lewis, Gilliland, and Bauer can be used to calculate bed [voidage Inglezakis and Pouloupoulos (15)]:

$$\varepsilon_f = \left[\frac{u_s}{u_{fm}} \right]^{1/m} \varepsilon \quad (16)$$

The exponent m is a function of the particle Reynolds number based on the minimum fluidization velocity. It can be estimated by the following correlation:

$$m = 4.21 \text{Re}_{fm}^{-0.0804} \quad (17)$$

The value of m is between 4.2 and 4.5 for $0.1 < \text{Re}_{fm} < 1$ and between 2.5 and 4.2 for $1 < \text{Re}_{fm} < 1000$.

Pavlov (1979) gave a simpler equation for ε_f based on the Archimedes number:

$$\varepsilon_f = \left(\frac{18 \text{Re}_p + 0.3 \text{Re}_p^2}{Ar} \right)^{0.21} \quad (18) \quad \text{Where} \quad Ar = \frac{d_p^3 \rho (\rho_p - \rho) g}{\mu^2} \quad (19)$$

MATERIALS AND METHODS

Resin Material

The ion-exchange resin employed was the OH^- type TULSION A-27 which was a strong base anion exchange resin. TULSION A-27 (OH^-) was obtained from the Thermax Company. The particles are in the shape of almost perfect spheres with an average diameter 0.7 mm (700 μm) and a wet density of 1.08 g/mL. The capacity of the resin was measured from the breakthrough curve of the $\text{OH}^-/\text{NO}_3^-$ exchange experiments. The total exchange capacity was about 1.2 meq/ml of resin.

Fluidized Bed System

The overall experimental apparatus is depicted in Figure 1. The column was filled with resin and washed with distilled water. Experiments were carried out in a glass column having 1 cm diameter and 100 cm high. To obtain the hydroxide, the resins were regenerated in downflow with four bed volumes of 4% NaOH solution and washed with distilled water. The temperature was maintained at 31 ± 1 °C. As the fluidized bed showed a quiescent behavior, the height of the bed could be determined visually.

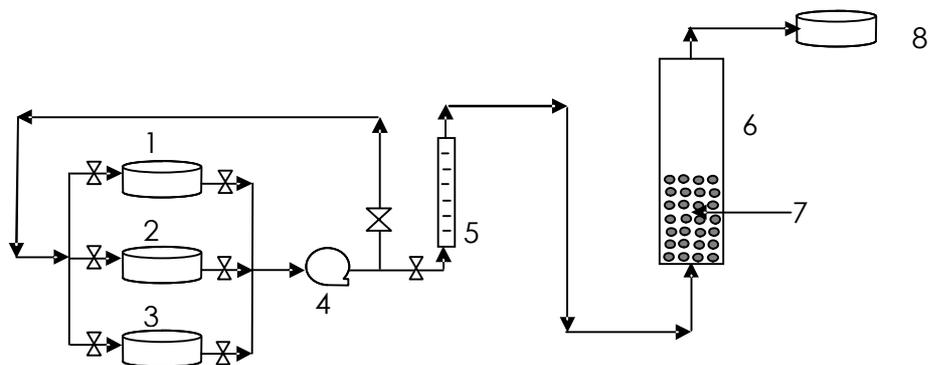


Figure 1. Experimental system: (1) NaNO_3 , (2) H_2O , (3) NaOH , (4) pump, (5) rotameter, (6) column, (7) resin, and (8) effluent.

CHEMICAL ANALYSIS

Nitrates were measured by a UV-Vis spectrophotometer. The absorbance was measured at 220 nm and a second reading was taken at 275 nm. This allowed correction for the interference due to dissolved organic matter. The difference between the two absorbance measurements was then calculated by the formula [Andre (18)]:

$$\text{Abs}_{220} - 2 \cdot (\text{Abs}_{275}) \quad (20)$$

RESULTS AND DISCUSSION

The effect of Reynolds number on fluidization voidage is presented in Figure 2. Values of the Richardson-Zaki coefficients n , and the apparent terminal velocity of the particle were calculated from linear regression of the plots of $\log(U)$ versus $\log(\epsilon)$, and the results are shown in Figure (3). The parameters correlated are listed in Table 1. An experimental value of $n=3.0007$ was obtained. The theoretical values of n were calculated by Equations 9, 10, and 15. A comparison of results showed a good agreement between the experimental and theoretical values of n and U_t . This comparison showed the operation of bed agreed with theory. The diffusion of nitrate ion with a resin particle at height 2 cm was slower than that at 3, 6, and 10 cm in the bed.

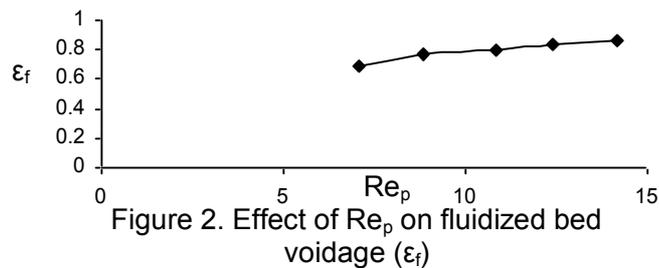


Figure 2. Effect of Re_p on fluidized bed voidage (ϵ_f)

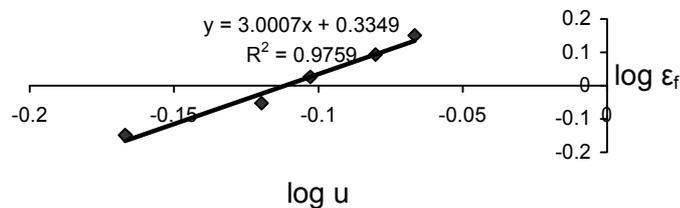


Figure 3. Richardson-zaki correlation between flow velocity and bed voidage for determination of n and U_t

Table 1. Experimental and theoretical values of U_t and n .

U_t (cm/s)		n			
Experimental	Eq. (7)	Experimental	Eq. (9)	Eq. (10)	Eq. (15)
2.162	2.16	3.0007	3.261	3.4	2.68

Figure (4) shows a comparison of the voidage estimated by various models at different particle Reynolds numbers. Figure (5) shows the experimental breakthrough curves at various bed heights.

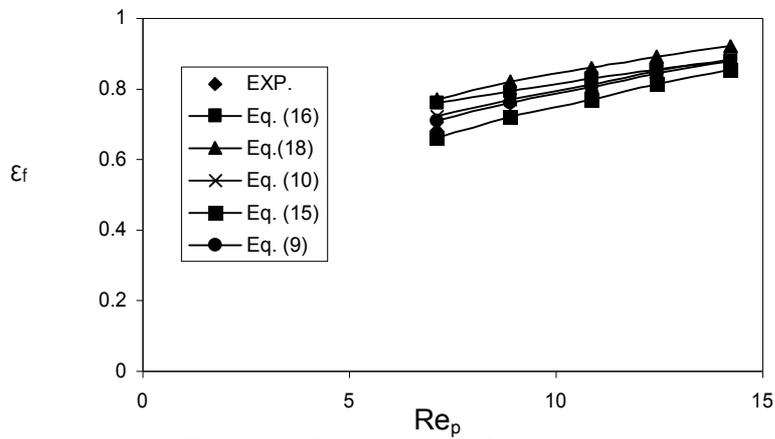


Figure 4. Comparison of overall voidage estimation models with experimental data

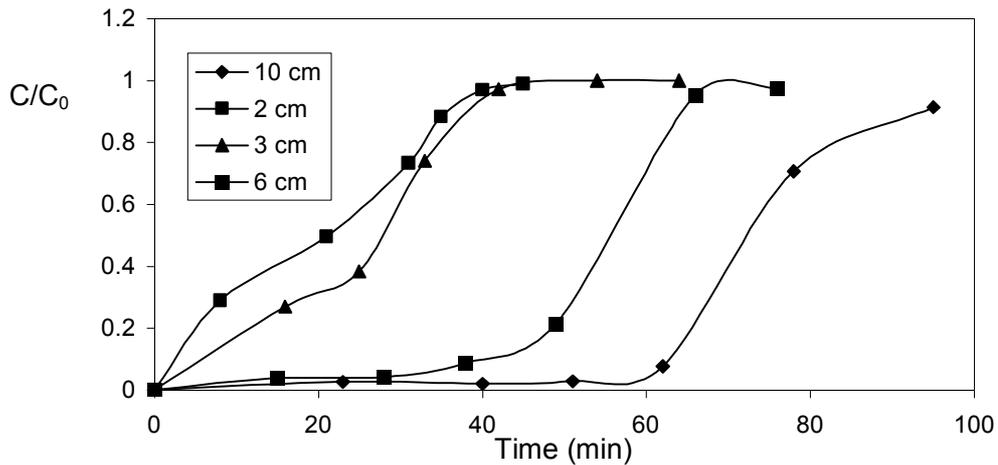


Figure 5. Breakthrough curves of NO_3 ions in the fluidized bed at different bed heights Z and a constant flow rate of 2 L/h

CONCLUSIONS

An experimental study of NO_3 removal from water by using a fluidized bed was carried out, and the following conclusion were made:

- 1- The Richardson and Zaki model showed a good fit with the experimental data, and can be use for voidage estimation.
- 2- Results of a fluidized bed operation using an ion exchange resin indicate that nitrate removal was improved significantly by increasing the bed height.

ACKNOWLEDGEMENT

The authors wish to acknowledge a research grant-in-aid from the Osmania University, Hyderabad, India.

NOTATION

Abs	absorbance measurements
Ar	Archimedes number
A_T	area of the cross-section of the column (m^2)
C	concentration (mol/m^3)
C_o	initial concentration (mol/m^3)
d_p	particle diameter (m)
g	gravity (m^2/s)
Ga	Gallileo number
G_a	Grashof number
H	bed height (m)
m_p	mass of particles (kg)
Re_{fm}	minimum fluidization Reynolds number
Re_p	particle Reynolds number
Re_t	terminal velocity Reynolds number
u_{fm}	minimum fluidization velocity (m/s)
u_s	superficial fluid velocity (m/s)
U	fluid velocity (m/s)
U_t	terminal fluid velocity (m/s)
V_L	liquid volume (m^3)
V_p	particle volume (m^3)
Z	bed height (m)
Z_f	bed height at fluidization (m)
ϵ	bed voidage
ϵ_f	voidage at fluidization.
ϵ_{fm}	voidage at minimum fluidization
μ	dynamic viscosity of the fluid (Kg/m.s)
ρ	density of the fluid (kg/m^3)
ρ_p	density of the particles (Kg/m^3)
Φ_s	the sphericity of the particle

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