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Modeling of zinc adsorption onto clinoptilolite in a slurry bubble column

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Authors (the order in which the authors appear - 1st, 2nd, etc. - is supposed to reflect their involvement in the research, the 1st author taking the lead)

HIGHLIGHTS

- Zinc is removed from Acid Rock Drainage by adsorption on clinoptilolite.
- Experiments were conducted in a slurry bubble column.
- Global kinetic model fits data well with mass transfer and uptake on adsorption sites.
- External mass transfer is not the rate-limiting step in this process.

An abstract summarizes all the sections of the paper. Abstracts are found before a full article in a journal, standalone in databases of abstracts, and in conference programs. The structure of an abstract and its length will depend on the journal or conference, as well as on the research field. Make sure you read their instructions to authors before you begin writing.

Keywords:
Zinc adsorption
Clinoptilolite
Slurry bubble column
Fluidized bed

Keywords are essential to get as many Google (Scholar) hits as possible and make the paper visible

abstract

This paper presents experimental results and a successful model for zinc removal from Acid Rock Drainage by adsorption on clinoptilolite. The experiments were conducted in a slurry bubble column of 0.09 m inner diameter and 1.4 m height, loaded with 100 g of clinoptilolite particles of diameter 0.3–1.4 mm/kg liquid. Aqueous zinc ion concentrations were determined before and during the adsorption tests. Zeolite was then regenerated by a sodium chloride solution, whose zinc concentration was determined against time. The Langmuir isotherm model was fitted to the experimental results under batch conditions to characterize the adsorption capacity of the solid surfaces. A global kinetic model with mass transfer to the particles and uptake on adsorption sites facilitates interpretation of the ARD slurry bubble column adsorption–desorption results. Due to air injection, perfect mixing could be assumed. The effect of particle size on the forward rate constant k_f of the adsorption process is important. The model is also applied to the sorbent regeneration by zinc desorption to provide a complete description. Consistent with the model, external mass transfer is not rate-limiting in this adsorption–desorption process.

1. Introduction

Acid Rock Drainage (ARD), generated naturally or from human activities (e.g. mining, highway construction), can cause serious environmental problems related to the significant increase in acidity and concentration of heavy metals in surface waters. Among metal ions, zinc is of concern due to its toxicity to the acquatic and terrestrial ecosystems. The most common treatment method for ARD sludges is to add limestone as a neutralization and precipitation agent. Due to the quantity of limestone required, and because it produces solid waste, this treatment can be expensive.

1) Setting the context/importance of research topic

Among the alternative sorbents for removing heavy metal ions, clinoptilolite, a natural zeolite, has proven to be promising (Doula et al., 2002; Lai, 2005; Petrus and Warchol, 2005; Dimirkou, 2007). Zeolites are efficient solid substrates for adsorb- ing aqueous ionic species in their extensive networks of channels and large specific internal surface area (Filippidis et al., 1996, Rivera et al., 2000; Li et al., 2008a, 2008b). Clinoptilolite is an inexpensive soil mineral with high adsorption capacity for zinc and copper (Doula et al., 2002; Dimirkou, 2007). Experiments carried out with packed beds (Lai, 2005) and rotating columns (Li, 2005; Li et al., 2005) confirmed high capacity for copper, zinc and aluminum. They also showed that clinoptilolite uptake/ removal of Zn occurred over 1–2 h, reaching 75% of capacity.

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An introduction typically follows different moves: 1) setting the context/establishing the relevance of the research topic; 2) showing there is some gap/limitation in existing research; 3) showing how this work resolves the gap/limitations or successfully extends or verifies past research

Introduction continued…

Reference for the paper as it would appear in a bibliography (only the title is missing). Note that only the 1st author's name appears in full, the other authors are conventionally referred to as "et al."

Preliminary studies (Cui et al., 2006; Li et al. 2008a, 2008b) demonstrated that a slurry bubble column (SBC) significantly enhanced mass transfer and sorption compared to traditional fixed beds and rotating columns.

1) Typical « review of previous work » phrase (preterit and/or present perfect)

Encouraged by these res[…] on clinoptilolite and in SB[…] 2010, 2012). Xu et al. (2[…] parameters on zinc immobilization: contact time, particle size, adsorbent dosage, initial pH and concentration. They showed that higher zinc uptake was obtained by increasing the initial zinc concentration, increasing pH and decreasing particle size. Xu et al. (2012) addressed the regeneration of the adsorbent by NaCl solution. They found that a go[…] g L^{-1} NaCl, an initial pH o[…] of 10 g g^{-1}.

2) Typical « research gap » phrase

To the best of our knowledge, no previous models have been proposed for cyclic zinc uptake/removal on/from clinoptilolite from zinc-spiked ARD. Menoud et al. (1998) proposed a model for the adsorption of heavy metal ions, including zinc, onto a chelating resin for two mixing limits: an ideal stirred tank model for CSTR experiments and a plug flow model with immobile solid for fluidized bed adsorption. Their global kinetic model considered mass transfer in a liquid film around the resin particles, intra- particle resistance to diffusion through the pores, and reaction on the adsorption sites. They found that external mass transfer was rate-limiting, both in CSTR and fluidized bed experiments. Homem et al. (2006) investigated nickel, zinc and lead removal by zeolite adsorption in a fluidized bed and simplified the model of Menoud et al. (1998) by assuming that the adsorption rate was limited by external mass transfer to the spherical particles.

The objectives of the present paper are twofold: first we propose a global kinetic model, with mass transfer around the solid particles and reaction on the adsorption sites, consistent with the results of our ARD slurry bubble column adsorption–desorption experiments. The effect of variation in the particle size is considered on the forward rate constant k_f. Secondly, based on the model, adsorption and mass transfer effects are assessed for the process. We then apply the same model to the regeneration of the sorbent by the desorption of metals with aqueous sodium chloride in order to model the entire adsorption–desorption process.

3) Typical « aim of the research » phrase

2. Material and methods

2.1. Sorbent

The sorbent for our experiments was clinoptilolite, a natural zeolite obtained from Bear River Zeolite, USA. No pre-treatment was required before use, except for oven-drying to equalize the moisture content of the various size fractions. The diameter of the clinoptilolite particles ranged between 0.30 and 1.40 mm. The mineralogical composition of the clinoptilolite was reported by Xu et al. (201[…] 3 and 0.72, […] adsorptio[…]

The typical functions of this section are:
-Provide qualitative and/or quantitative details about the approach taken
-Present tools used in the study
-Refer to sources of information and protocols
Here, both mathematical models and experimental approaches are presented.

2.2. Sorb[…]

ARD from Highway 97C near Merritt, British Columbia with a pH of 3.2 was used in the experiments. The concentration of the main metals is reported in Table 1. Zinc, our primary focus, is the contaminant of major concern in this ARD.

Table 1
Concentrations of major metals in ARD […] Merritt, British Columbia.

Units	Metal concentrations						
	Zn	Mn	Al	Ca	K	Na	Si
mg/kg	14.4	10.6	21.9	198	4.5	179	40.4
mmol/kg	0.220	0.193	0.812	4.94	0.115	7.79	1.44

2.3. Slurry bubble column

The experiments were conducted in a slurry bubble column of 0.09 m inner diameter and 1.4 m height. Based on previous work (Chen, 2005; Cui et al., 2006; Lai, 2005; Xu et al., 2010, 2012), the slurry was prepared to provide 100 g clinoptilolite/kg liquid, with 2 kg ARD in each experimental run. Air was introduced into the bottom of the column through a calming zone before passing through a perforated plate containing 57 uniformly-distributed orifices of diameter 6.3 mm, with a screen underneath to prevent particles from passing. A constant superficial gas velocity of 0.08 m/s was sufficient to create strong internal liquid circulation in the column, thereby ensuring nearly perfect mixing. The gas holdup was determined by the direct reading of the bed height (H) under aerated conditions with H ¼ 0.425 m.

2.4. Adsorption equilibrium experiments

In these experiments, 200 g of dry clinoptilolite and 2 kg of ARD solution with an initial zinc concentration, C_0, of 1.53 mol/m^3, were loaded into the column and stirred at a constant temperature of 15 1C for 1 h. Time 0 is taken immediately after introducing the solids into the column. The supernatants of equilibrate solutions were analysed to determine the equilibrium zinc concentration, C_e. Zinc concentrations were determined by an Atomic Adsorption Spectrophotometer (AAS) and an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES). The equilibrium zinc uptake, q_e, on the clinoptilolite was calculated by a mass balance.

The passive voice is very often used to describe actions (here, the different stages of an experiment)

2.5. Adsorption kinetic experiments

In the kinetic experiments, the ini[…] to that used in equilibrium experi[…] differs. Experiments were stopped at different time (5, 10, 15, 20, 25, 30, 40, 60 min). After contacting for the designated time, the supernatant liquid was rapidly separated from the clinoptilolite samples by filtration. Metal ion concentrations of the supernatant were determined by an Atomic Adsorption Spectrophotometer (AAS) (Thermo Jarrell Ash Video 22, TJA Solutions, US). Using this procedure, the dynamic evolution […] without sampling.

You should check for specific in-text citation conventions, depending on the subject and the journal (here, Author, year)

2.6. Regeneration experiments

As in previous studies (Chen, 2005; Li et al., 2005; Semmens and Martin, 1988, Xu et al., 2010, 2012), NaCl acted as the regenerant. In a recent paper (Xu et al., 2012), a 20 g L^{-1} NaCl concentration, an initial pH of 3 and a regenerant/sorbent mass ratio of 10 provided the best operating conditions and were therefore adopted in the present work. Regeneration was undertaken by re-suspending 200 g of previously-used clinoptilolite particles with 20 g/L aqueous NaCl solution in the column by an upward air flow at the same temperature and superficial velocities as in the adsorption tests for pre-specified time periods to determine the rate of release of zinc as a function of time.

These studies were also carried out in the batch mode using the slurry bubble column.

3. Results

The main functions of the results section include:
-presenting original data
-referring to figures, equations and tables
-describing control samples or groups
-presenting conclusions
It is a key section where researchers show how they have contributed to advancing knowledge in a specific scientific area.

3.1. Adsorption equilibrium study

The Langmuir model (E… equilibrium isotherms in or… capacity q_m and dissociation…

$$\frac{q_e}{q_m} = \frac{C_e}{K_d + C_e}$$

with the dissociation constant $K_d = k_r/k_f$ where k_r and k_f represent the reverse and forward rate constants respectively. C_e is the equilibrium zinc concentration (mol/m^3), q_e is the zinc mole adsorbed per unit mass at equilibrium (mol/kg) and q_m is the maximum clinoptilolite capacity (mol/kg). Fig. 1 shows an example of the fitting for the particle size range 0.30–0.40 mm.

The Langmuir isotherm characteristics obtained for the four different cuts of solids are reported in Table 2. The maximum resin capacity q_m decreased from 10.5 to 7.0 with increasing particle diameter, whereas the dissociation constant smoothly decreased from 0.785 to 0.641 with increasing particle size. This adsorption capacity is greater for finer particles because of better access to the exchange sites.

3.2. Adsorption kinetics

Fig. 2 reports the experimental time variation of zinc concentration on clinoptilolite for different particle sizes. Whatever the particle size, a strong decrease in zinc concentration was observed during the initial period of the adsorption process, followed by a more gradual decrease versus time. Consistent with Table 2, greater zinc uptake was observed with the smaller particles.

Figures and tables must always be referred to and explained very clearly in the body of the paper

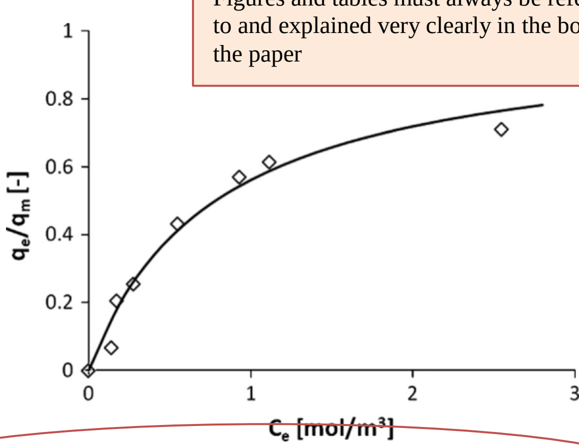


Fig. 1. Example of the equilibrium isotherm for zinc uptake on clinoptilolite. Particle size: 0.30–0.50 mm.

3.3. Modeling

3.3.1. Main assumptions and equations

As in the analysis proposed by Menoud et al. (1998), the adsorption of zinc onto zeolite is represented by a second order reversible interaction:

$$S + Zn^{2+} \rightleftharpoons SZn^{2+}$$

… S is a free site and SZn^{2+} is an occupied one.

…e rate of adsorption is given by

$$\ldots C_i (q_m - q) - k_r q \quad (2)$$

where C_i and q are the zinc concentrations inside the pores and on the clinoptilolit… Langmuir isoth… Experiment… bubble column… transfer resista… adsorption–des… and their densi…

Information is often organized in short paragraphs. Paragraph structure follows simple rules: 1) it begins with a topic sentence, then the rest of the paragraph is used to develop this topic; 2) sometimes there is a short concluding sentence at the end of the paragraph; 3) information is given from the general to the increasingly specific; 4) information is given in the most logical and consistent order.

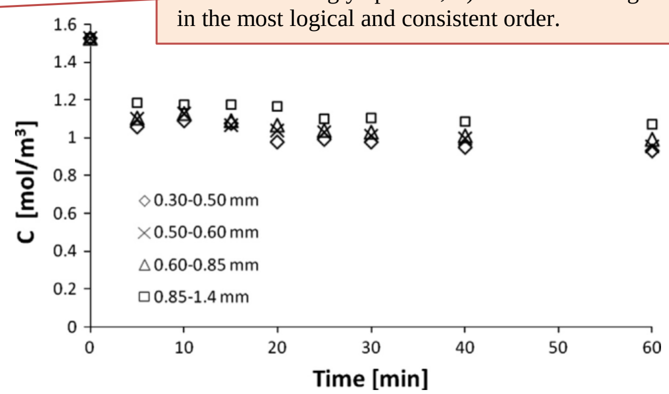


Fig. 2. Zinc adsorption versus time for different particle sizes in a slurry bubble column.

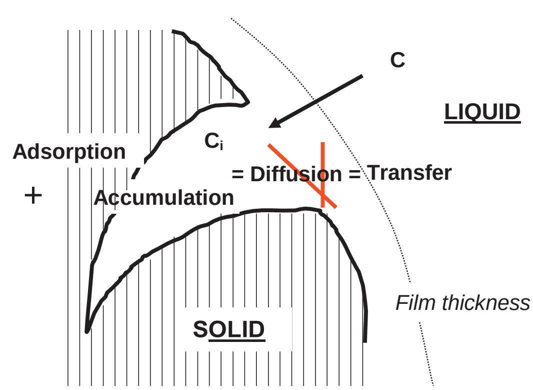


Fig. 3. Schematic description of the model assumption (with negligible diffusion limitation inside the pores).

In a scientific paper, all the tables and figures must have a clear title and be numbered

Table 2
Langmuir isotherm characteristics for zinc adsorption on clinoptilolite at 20 °C.

Particle size range [mm]	0.30–0.50	0.50–0.60	0.60–0.85	0.85–1.4
Fitted dissociation constant, K_d [mol/m^3]	0.785	0.771	0.678	0.641
Maximum clinoptilolite capacity, q_m [mol/kg] 10^3	10.49	9.47	8.17	6.98

Table 3
k_L from Eq. (7) and best-fit values of parameter, k_f.

Particle size range [mm]	0.30–0.50	0.50–0.60	0.60–0.85	0.85–1.4
Average particle size [mm]	0.40	0.55	0.73	1.13
Liquid–solid mass transfer coeffcient, k_L [m min^{-1}]	0.0104	0.0098	0.0093	0.0086
Forward rate constant, k_f [m^3 mol^{-1} min^{-1}]	0.0907	0.0535	0.0506	0.0545

are written for batch conditions:

$$\frac{dC}{dt} = -k_L a (C - C_i) \tag{3}$$

$$\frac{dC_i}{dt} = \frac{k_L a \varepsilon_{l,ext}}{\varepsilon_{l,in}}(C - C_i) - \frac{\rho_m \varepsilon_s}{\varepsilon_{l,in}}\left[k_f C_i (q_m - q) - K_d q\right] \tag{4}$$

$$q \varepsilon_s \rho_m + C \varepsilon_{l,ext} + C_i \varepsilon_{l,int} = C_0 (\varepsilon_{l,int} + \varepsilon_{l,ext}) + q_0 \varepsilon_s \rho_m \tag{5}$$

$$a = \frac{6\varepsilon_s}{d \varepsilon_{l,ext}} \tag{6}$$

C and C_i are the concentrations in the bulk and inside the pores respectively, k_L is the liquid-side mass transfer coefficient, a is the interfacial area and ε defines the different volume fractions (see Nomenclature). This approach, shown in Fig. 3, neglects any diffusion limitation inside the pores, and assumes that the global kinetics are mainly regulated by the adsorption on the internal sites and/or by external mass transfer.

The liquid–solid mass transfer coefficient in a slurry bubble column has been estimated from the correlation proposed by Kikuchi et al. (1998) for the three-phase flow which relates the Sherwood number (Sh) to the Schmidt number (Sc) and the energy dissipation rate, E:

$$Sh = 2 + 0.47 \left(\frac{E^{1/3} d^{4/3}}{\nu_L}\right)^{0.63} Sc^{1/3} \tag{7}$$

$$E = \left[(\rho_s - \rho_g)\varepsilon_s + (\rho_l - \rho_g)\varepsilon_l\right] u_g g / (\rho_l \varepsilon_l) \tag{8}$$

Eq. (7) was applied with a 11% standard deviation to the data of Kikuchi et al. (1995) obtained in a slurry bubble column of 0.12 m i.d. and for cation exchange resin beads of size comparable to the solids in the present study. Neglecting the effect of other ionic species, the diffusion coefficient of zinc in water was set equal to 2.3×10^{-9} m^2/s, from Weingärtner (1982).

After estimating k_L from Eq. (7), the only unknown parameter in Eqs. (3)–(6) is k_f, whose value is fitted by minimizing the difference between model predictions and the experimental data for both the adsorption and desorption processes.

Eqs. (3)–(6) can be solved with the initial condition:

at $t = 0$, $C_i = 0$ and $C = C_0$

However, to respect the condition that $q = 0$ at $t = 0$, the liquid volume fractions inside and outside the particles are

at $t = 0$, $\varepsilon_{l,int} = 0$ and $\varepsilon_{l,ext} = \dfrac{m_L}{\rho_L A H}$

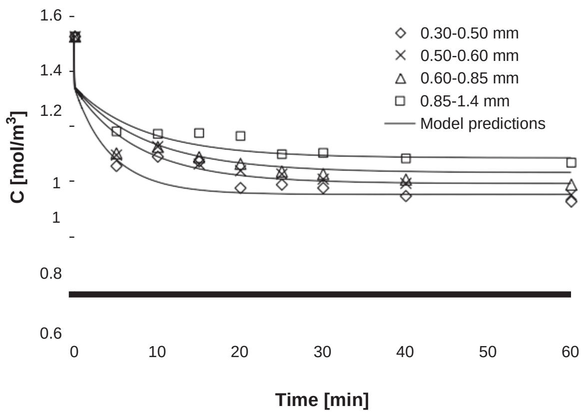


Fig. 4. Experimental and predicted zinc concentrations versus time for the adsorption in slurry bubble column with different particle sizes.

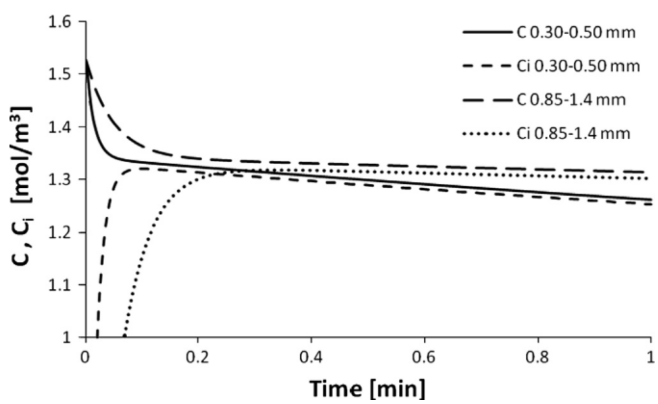
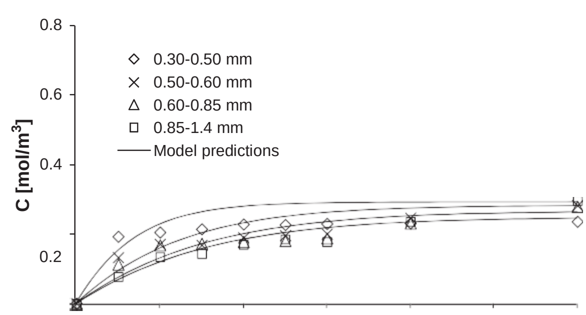


Fig. 5. Zinc concentration in the bulk, C, and at particle surface, C_i, during adsorption predicted by the model.



This paper has no "Discussion" section, which usually appears after the Results section. This section typically includes:
-a recap of the main findings
-an introduction to the limitations of the study
-a description of key unanswered questions
-potential future avenues of investigation

4. Conclusions

A simple global kinetic model is proposed, consistent with the ARD zinc uptake and the removal experiment in a slurry bubble column. The Langmuir isotherm model is applied to determine the maximum solid capacity, q_m, and the dissociation constant K_d. The maximum sorption capacity was obtained with the smallest particles (0.30–0.50 mm) tested. Zinc adsorption kinetics on clinoptilolite showed greater uptake with the smallest particles. Based on mass transfer around the solid particles and uptake on the adsorption sites, the proposed model predicts zinc concentra- tions as a function of contact time well for both adsorption and desorption, for a wide range of particle sizes. Moreover, there was little difference observed between the external and internal zinc concentrations according to the model, showing that internal mass transfer is not the rate-limiting step in the process and that it is not necessary to account for internal diffusion limitations through the pores of the clinoptilolite particles.

The Conclusion section typically reminds the paper's objective and focus, and recaps the main hypotheses and findings

Nomenclature

A column cross-sectional area (m^2)
a interfacial liquid–solid surface area (m^2/m^3)

C aqueous zinc concentration in bulk (mol/m^3)
C_e aqueous zinc concentration at equilibrium (mol/m^3)
C_{exp} experimentally measured concentration (mol/m^3)
C_i aqueous zinc concentration inside pores (mol/m^3)
C_{model} concentration predicted by model (mol/m^3)
D diffusion coefficient (m^2/s)
d particle diameter (m)
E energy dissipation rate per unit mass of liquid (m^2/s^3)
H total bed height (m)
i integer (dimensionless)
K_d dissociation constant (mol/m^3)
k_f foward rate constant ($m^3 mol^{-1} min^{-1}$)
k_r reverse rate constant ($m^3 mol^{-1} min^{-1}$)
k_L liquid side mass transfer coefficient ($m\ s^{-1}$)
m_L total liquid mass (kg)
N number of experimental determinations (dimensionless)
q zinc mole adsorbed per unit mass (mol/kg)
q_0 zinc mole adsorbed per unit mass at t¼ 0 (mol/kg)
q_e zinc mole adsorbed per unit mass at equilibrium (mol/kg)
q_m maximum clinoptilolite capacity (mol/kg)
Sh Sherwood number for three-phase reactor ($k_L d/D$) (dimensionless)
Sc Schmidt number (n_L/D) (dimensionless)
e_s solid fraction $m_s/AH/r_m$ (dimensionless)
$e_{l,ext}$ external liquid fraction (dimensionless)
$e_{l,int}$ internal liquid fraction (dimensionless)
e_g gas fraction,¼ 1—e_s—$e_{l,ext}$—$e_{l,int}$ (dimensionless)
e_m clinoptilolite mass density (kg/m^3)
n_L kinematic viscosity (m^2/s)

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