Journal of Pollution Effects & Control

Abdullah, J Pollut Eff Cont 2014, 2:2
DOI: 10.4172/2375-4397.1000120

Research Article

Open Access

Aluminum Pollution Removal from Water Using a Natural Zeolite

Ali M Abdullah*

Holding Company for Water and Wastewater, Cairo, Egypt

Abstract

Aluminum ion adsorption capacity of zeolite from polluted tap water was investigated in this study. For the experimental studies, zeolite mineral which was 20-200 mesh size was used by activating with HCl and aluminum ion removal efficiencies were examined. Experiments were studied under laboratory, batch conditions were run at different pH values, temperatures. The highest removal efficiency was found as about 84.7% at pH 6.5. Similarly, experiments were carried out at different temperature values, and the maximum efficiency was obtained at 30°C. The efficiency obtained under the same conditions was 93%. The highest aluminum removal efficiency was obtained with 180 rpm shaking speed.

Keywords: Aluminum removal; Water; Adsorption; Zeolite

Introduction

Aluminum can occur in a number of different forms in water. It can form monomeric and polymeric hydroxy species, colloidal polymeric solutions and gels, and precipitates, all based on aquated positive ions or hydroxylated aluminates. In addition, it can form complexes with various organic compounds (e.g. humic or fulvic acids) and inorganic ligands (e.g. fluoride, chloride, and sulfate), most but not all of which are soluble. The chemistry of aluminum in water is complex, and many chemical parameters, including pH, determine which aluminum species are present in aqueous solutions. In pure water, aluminum has a minimum solubility in the pH range 5.5-6.0; concentrations of total dissolved aluminum increase at higher and lower pH values.

The concentration of aluminum in natural waters can vary significantly depending on various physicochemical and mineralogical factors. Dissolved aluminum concentrations in waters with near-neutral pH values usually range from 0.001 to 0.05 mg/l but rise to 0.5-1 mg/l in more acidic waters or water rich in organic matter. At the extreme acidity of waters affected by acid mine drainage, dissolved aluminum concentrations of up to 90 mg/l have been measured [1].

The presence of Al in drinking water has given rise to discussions on possible of risks, because of its suspected connection with Alzheimer's diseases or dialysis encephalopathy [1]. Driscoll and Letterman [2] reported that dialysis patients exposed to elevated Al may exhibit dialysis enchepholopathy, and/or bone mineralization disorders such as dialysis osteodystrophy. Martyn et al. [3] based on a survey of 88 county districts in England and Wales reported that rate of Alzheimer's disease was 1.5 times higher in districts where the mean Al concentration exceeded 0.11 mg/l than in districts where concentrations were less than 0.01 mg/l. Al shows low acute toxicity. Chronic exposure data are limited, but indicate that Al likely interferes with phosphorus absorption, and causes weakness, bone pain and anorexia.

The most widely used methods for removing metal ions are: precipitation with chemical and electrochemical methods [4]. A major problem with this type of treatment is the disposal of the precipitated wastes. Ion exchange technique which the second most widely used method for metal ion removal does not present a sludge disposal problem and has the advantage of reclamation of Al (III) [5]. This method can reduce heavy metals to very low levels. However, ion exchange treatment does not appear to be economical. Activated carbon is also efficient for removal of trace elements from the waste water, but its high cost when usage. The adsorption phenomenon has still been found economically appealing for the removal of toxic metals from water by choosing some adsorbents under optimum operation conditions.

It has been reported that some aquatic plants [6], agricultural by-products [7,8], sawdust [9], clay [10], zeolite, turba [11] and microorganisms have the capacity to adsorb and accumulate heavy metals. Cost comparisons are difficult to make due to the scarcity of consistent cost information. Although many experiments have been accomplished in the area of low-cost sorbents, a great deal of work is necessary to understand better low-cost adsorption processes and to demonstrate the technology.

Natural zeolites have ion exchange and removal capacity. zeolites has high removal capacity of metal ions Pb^{+2}, Cu^{+2}, Zn^{+2}, Cd^{+2}, Ni^{+2}, Al^{+3}, Fe^{+2} and Mn^{+2}. It is an economical ion exchange material comparing with the synthetic resin.

Natural zeolites have been used as adsorbent and ion exchanger with discovering crystal feature and some chemical structure. It is physical structure look like a selective sieve and named as molecular sieve by Mc Bain.

General formula of the Zeolites is $X[(M^{+}_{1}, M^{++}_{1/2}).(AlO_2)].ySiO_2.zH_2O$. M is a cation ($Na^+$, K^+, Ca^{++}, Pb etc.) which ratio (y/x) changes from 1 to 5. M^{++} is a cation (Ca^{++}, Mg^{++} and Ba^{++} etc.) change with M^{+}_{1}. SiO_2/AlO_2 molar ratio or (y/x) zeolite changes with zeolite types from 1 to 5. SiO_4 and AlO_4 octahedrals are the monomers of the zeolites that Si or Al was centre of the structure and oxygen was on the corners. Natural zeolites were also uses at industrial fish ponds [12]. Chelischev [13] used zeolites as ion exchange for heavy metals. Similarly Sato and Fukagawa, used zeolites for nitrogen removal at detergent industry waste water.

The aim of this study was removal of the ionic aluminum pollution

---

***Corresponding author:** Ali Mahmoud Ahmed Abdullah, Ph. D, Technical Manager at Holding Company for Water and Wastewater, Cairo, Egypt, Tel: 002-01229248037; E-mail: dr2252000@dr.com

Received September 19, 2014; **Accepted** October 06, 2014; **Published** October 16, 2014

Citation: Abdullah AM (2014) Aluminum Pollution Removal from Water Using a Natural Zeolite. J Pollut Eff Cont 2: 120 doi: 10.4172/2375-4397.1000120

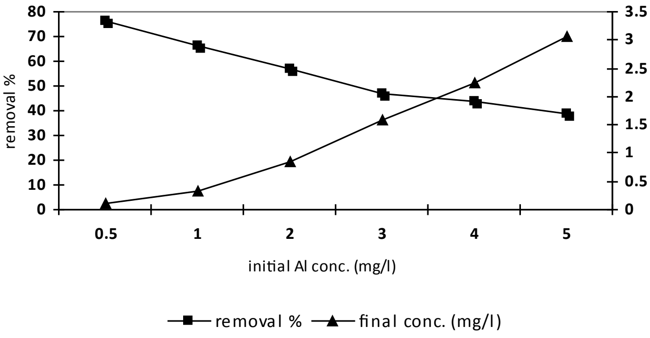

Figure 1: Al^{+3} removal efficiency of zeolite from aqueous solutions containing different Al^{+3} levels.

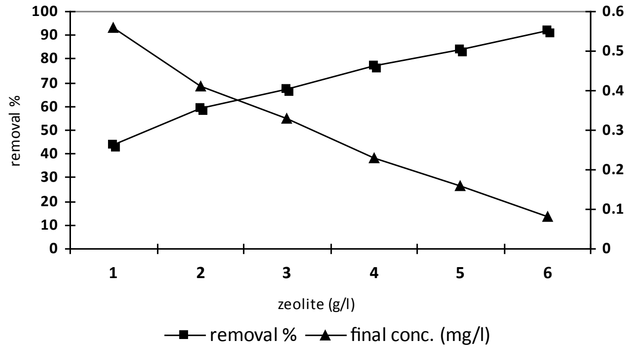

Figure 2: Effect of different adsorbent dose of zeolite for Al^{+3} adsorption.

from the preliminary treated water using zeolite as a natural and economical material.

Materials and Method

- Zeolite used provided from Iran (Chem-lab, Co., Cairo, Egypt).
- The chemical analysis showed that, the zeolite was in hydrated alumina-silicate mineral containing mostly alkali and soil alkali metals originated in holes of volcanic rocks. They contain hydrate in holes connected each other. Particle size is 50-2000 µm of 82.56% materials and 2-50 µm others. Porosity ratio was about 50% and natural hydrate capacity and ion exchange capacity were 25.68% and 71.73 meq/100 g respectively.
- Materials used in this study were between 20-200 mesh sizes.
- *Gallenkap* (U.K.) thermostatic shaker incubator was use for batch experiments.
- The concentration of Al used in the present studies varies from 0.5 to 5 mg/l.
- Al concentrations residues were measured by Dr. Lange Cadas-200 Spectrophotometer (Germany).
- The pH was adjusted to required values using *Jenway 3010* pH meter.
- *Gec-Avery VA/WA* Analytical Balances was used to weighing procedure for preparation of solution.
- Zeolite in 20-200 mesh size was treated with acid or base solution in different concentration to improve its adsorption capacity. For this process, 2.0 grams of zeolite in 100 ml distilled water containing different amounts of acid or base was shaken at different shaking seeped and temperature, and finally 1 h 200 rpm and 23 h 100 rpm at 30°C stable temperature.
- Zeolite samples were washed twice 2 with distilled water after treating aside or alkali solution. With this procedure, the best activation media was determined 2.0 ml 1 M HCl additions. For this reason, activated zeolite samples with 2.0 ml 1 M HCl were used in following experiments. After designing the activation media, to optimum adsorbent amount, each 2 grams of activated zeolite was added in 100 ml tap water containing different concentration of Al^{+3}.
- After 1 h contact time in batch rector, zeolite was filtered with fine nylon filter and Al^{+3} concentrations were measured using Cuvette test via Cadas 200 UV -Vis spectrophotometer.
- Effect of pH on the adsorption capacity of 2 grams of zeolite was tested adjusting pH of the 100 ml sample solution (containing 40 mg-Al/l) via adding different amount of acid or base. Effect of ambience temperature was also investigated.
- Experiments were run at the temperatures of 10, 20, 30, 40 and 50°C the conditions designated earlier experiments.

Results

Zeolite samples were activated using 1 M HCl or NaOH to increase the adsorption capacity. For this purpose, firstly 1, 2 and 3 ml 1M HCl were added in three conical flask containing 2 grams of zeolite and 100 ml distilled water separately, secondly 1, 2 and 4 ml NaOH were added in an other conical flask containing 2 grams of zeolite separately, and one conical flask containing 2 grams of zeolite was kept without acid or alkali addition. Results showed that the best activation condition was addition of 2.0 ml 1 M HCl in100 ml distilled water containing 2 grams of zeolite.

Effect of initial Al (III) concentration by the zeolite adsorption

Determination of effective Al^{+3} concentrations on removal efficiency of zeolite was investigated with addition of 2.0 grams of activated adsorbent in different concentration of Al^{+3} in 100 ml water solutions. Figure 1 shows that the maximum removal efficiency was obtained with 1.6 mg/L Al^{+3} concentrations, a little decrease was found below from this values. Later on, decreasing removal efficiency was detected with increasing initial heavy metal concentrations in the solutions. It is likely that a given mass of adsorbent material has a finite number of adsorption sites, and that as metal concentrations increase, these sites become saturated. That is, there is some metal concentration that produces the maximum adsorption for a given adsorbent mass, and there after, adding more metal cannot increase adsorption because no more sites are available: all are occupied [7].

Effect of adsorbent doses on the Al^{+3} adsorption by the zeolite was investigated with changed adsorbent doses from 1.0 to 4.0 g that maximum removal efficiencies have been achieved to be 67% with 2.0 g zeolites /100 ml solution (Figure 2). Residual Al^{+3} concentration seemed until to 2.0 g a clear increasing and than a smaller increasing after this value.

pH effect on adsorption capacity

Effect of pH on the adsorption of Al^{+3} by the zeolite was also investigated with the changing pH values from 3.0 to 9.0. The results are depicting the dependence of the Al^{+3} removal as a function of pH

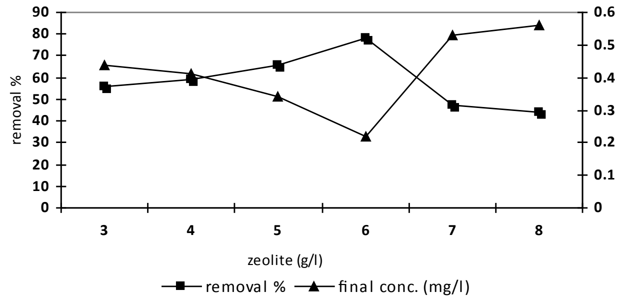

Figure 3: Effect of solution pH on removal efficiency of Al^{+3} from water.

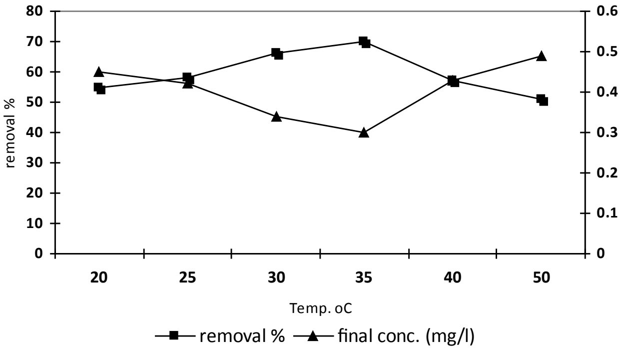

Figure 4: Effect of sample temperature on the zeolite removal efficiency of Al^{+3} from the water.

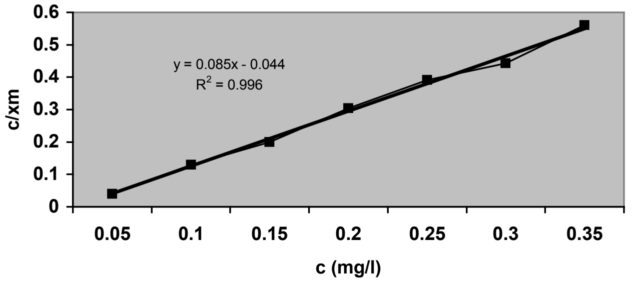

Figure 5: Langmuir adsorption isotherm plot for Al^{+3} adsorption.

showed that maximum removal efficiencies have been achieved to be 85% at pH 6.0 (Figure 3). According to different pH values Al ions may present in different forms. For this reason, Al^{+3} ions dominates at pH<7 and $Al(OH)_3$ dominates at pH>7 [8].

Effect of sample temperature on Al^{+3} removal capacity of zeolites

Activated 2 grams of zeolite in 100 ml water at pH 6.0 was tested for adsorption of Al^{+3} at different sample temperature between 10 to 50°C. Results showed that optimum working temperature was about 30°C (Figure 4). Removal of Al^{+3} from the water was 88% at this temperature; it was lowered at lower and at higher temperatures.

The equilibrium adsorption isotherm for Al^{+3} on activated zeolite was plotted for varying adsorbent doses changing from 0.25 to 3.0 g L^{-1}) and fixed initial Al^{+3} concentration (1.6 mg L^{-1}) at pH 6.0. The percentage of Al^{+3} adsorption decreased with increasing adsorbate amount, which indicates that the adsorption depends upon the availability of the binding sites for Al^{+3}. In order to determine the adsorption capacity of the activated zeolites, the equilibrium data for the adsorption of Al^{+3} were analyzed in the light of adsorption isotherm models. Mostly used adsorption isotherms are Langmuir and Freundlich equations which were given below respectively (Equation 1 and 2):

$$(x/m) = (1/a) + (1/ab).\ (1/c) \tag{1}$$

$$\mathrm{Ln}\ (x/m) = \mathrm{Ln}\ (k) + (1/n)\ .\ \mathrm{Ln}\ (c) \tag{2}$$

where x/m is the amount of Al^{+3} adsorbed from per unit weight of adsorbent (mg g^{-1}), c is the concentration of Al^{+3} at equilibrium (mg L^{-1}), $1/ab$ is slope for plot with $1/a$ contact point on y axis for Lanmuir adsorption isotherm, and $1/n$ is slope for plot with contact point of $\ln$ k constant on crossing point of y axis for Freundlich adsorption isotherm. The straight line nature of the graph indicates that the adsorption confirms the Langmuir model. The experimental data points were fitted to the Langmuir equation (Figure 5), but it was not fit well to the Freundlich equation. Chemical kinetics of Al^{+3} removal was also investigated in batch reactor for 60 min. contact time that checked with °0, °1, °2 and °3 order reaction equations. Only our date is quite well fit to 2nd order reaction kinetic (Figure 6). Square of correlation coefficient was R^2=0.9905 which confirms good relation between contact time and concentration change by experimental period. Reaction kinetic of Al^{+3} removal from the aqueous environment by zeoliteis 2nd order that adsorption of zeolite was physical character and it is suitable for reuse.

Discussions

This paper presents the results of Al^{+3} ions removal from aqueous solution using common, naturally occurring zeolites. Operational parameters such as initial Al^{+3} concentrations, adsorbent doses, pH of the solution, temperature, and contact time clearly affect the removal efficiency. The optimum Al^{+3} removals by the zeolite were obtained at pH 6. At pH higher than 7, Al^{+3} removal probably resulted primarily from precipitation rather than adsorption. According to results reported in this study, Al^{+3} adsorption mechanisms by zeolite may be explained as follows:

- Vander-walls and hydrogen binding of Al^{+3} with hydroxyl group of zeolite surfaces.
- Ion exchange between Al^{+3} ions and exchangeable cations (Na, K etc.).
- Diffusions of Al^{+3} through selective sieve of zeolite named as molecular sieve (Mc Bain, 1932).

According to these results, natural zeolite appeared to be a promising

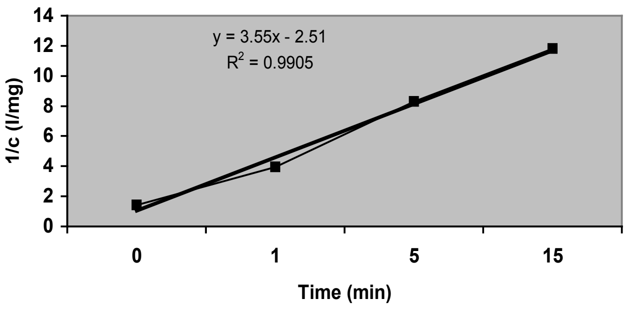

Figure 6: Reaction kinetic (2nd order) plot for the Al^{+3} adsorption of activated zeolite (1.6 mg L Al^{+3} concentration and 15 min contact time period).

adsorbent for the removal of the Al^{+3} from the polluted water. A process using the zeolite for the removal of heavy metal seemed to be potentially more economical than current process technology. However, further studies would suggest, since different metals and different adsorbents are known to have different properties structures.

Heavy metals affect to human health because of their harmful effect. Nevertheless, a lot of heavy metals were using in the industry and they join to food chain. In this respect, wastewaters coming from pollution sources must be treated to decrease their concentration down to limits given in water regulations before to give in the environment. Main principle of removal processes base to chemical precipitation of metal ions as hydroxide. Adsorption, ion exchange, membrane filtration and reverse osmosis etc. were used for advanced treatment. Removal of the metal pollution from the water with the classical methods was not sufficient, practical and economical. Ion exchange method may be used after pre-treatment operation. Most of the heavy metals recovery was not easy from the precipitate and economic. For this reason, adsorption processes by the natural zeolites may be preferred in the application.

In this investigation, a natural zeolite, as an adsorbent was used for removal of Al^{+3} from the water and very high removal efficiencies were found with different working conditions. Zeolite was activated treating with different concentrations of acid and alkali solution in the first step. Tests showed that optimum activation condition was addition of 2 ml 1 M HCl in 100 ml water containing 2 grams of zeolites.

After activation process, other experimental conditions were investigated. The second test was determination of usage of concentration of Al^{+3} which was optimum at 1.6 mg L^{-1}. Removal rate was reached about 66% with this Al^{+3} concentrations. In a separate investigation, with activated zeolite at same condition given above and 40 mg L^{-1} Al^{+3} concentrations adsorption solution was optimum condition.

pH of the adsorption solution was other important condition which was tested. A maximum removal efficiency of 85% was found at pH 6. This showed that zeolite was activated in acidic condition changing cations with H^+ ions and Al^{+3} ions form in the working solution. After this experiment temperature and contact time were determined. Removal efficiency was increased until 30°C, and then decreased.

All these experimental results showed that zeolite is suitable adsorbent for removal of heavy metals. Zeolite is available in economical price. It is suitable material for heavy metal removal from the industrial waste water.

References

1. Jekel MR (2005) Aluminum in water: How it can be removed? Use of aluminum salts in treatment. Proc. of the Int. Water Supply Ass., Copenhagen, Denmark, May 25-31.
2. Driscoll CT, Letterman RD (2005) Chemistry and fate of Al III in treated drinking water. J. Environ. Eng. Div., ACSE 114: 21-37.
3. Martyn CN, Osmand C, Edwardson JA, Barker DJ, Harris EC, et al. (2008) Geographical relation between Alzheimer's disease and aluminum in drinking water. The Lancet. 1: 59-62.
4. Chen G (2004) Electrochemical technologies in wastewater treatment Separation and Purification Technology 38: 11-41.
5. Petrus R, Warchol J (2003) Ion exchange equilibria between clinoptilolite and aqueous solutions of Na^+/Cu^{2+}, Na^+/Cd^{2+} and Na^+/Pb^{2+}. Microporous and Mesoporous Materials.
6. Axtell NR, Sternberg SP, Claussen K. (2003) Lead and nickel removal using Microspora and Lemna minor. Bioresour. Technol. 89: 41-48.
7. Argun ME, Dursun Ş, Özdemir C, Karataş M (2007a) Heavy metal adsorption by modified oak sawdust: thermodynamics and kinetics. J Hazard. Materials. In Press.
8. Argun ME, Dursun S, Gür K, Özdemir C, Karatas M, et al. (2005) Adsorption of copper on the modified wood (pine) materials. Cell. Chem. Technol. 39: 581-593.
9. Özdemir C (2005) Chromium removal from the wastewater on raw material of saw dust. Cell Chem. Technol. 39: 325-329.
10. Márquez GE, Ribeiro MJP, Ventura JM, Labrincha JA (2004) Removal of nickel from aqueous solutions by clay-based beds. Ceramics International 30: 111-119.
11. Ho YS, Wase DAJ, Forster CF (1995) Batch nickel removal from aqueous solution by sphagnum moss peat. Water Res. 29: 1327-1332.
12. Ames LL (1967) Zeolitic Removal of Ammonium Ions from Agricultural Waste Waters, Proc. 13th Pasific Northwest Ind. Waste Conf, Washington State Univ, 135-152.
13. Chelischev NF (1974) Ion Exchange of Heavy Metals on Clinoptilolite, Doc. Acad Nauk. S.S.S.R. 217: 1140-1141.