CHEMICAL ENGINEERING TRANSACTIONS
VOL. 46, 2015
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.aidic.it/cet
Guest Editors: Peiyu Ren, Yancang Li, Huiping Song

ISBN 978-88-95608-37-2; **ISSN** 2283-9216
DOI: 10.3303/CET1546014

Experimental Study on Ammonia Nitrogen Adsorption Performance of Zeolite Powder

Jiaqi Wang[a], Wenbiao Jin[a], Hua Guo[b], Xinyue Wang[b], Junliang Liu*[b]

[a] Harbin Institute of Techology Shenzhen Graduate School, Shenzhen 518000, China,
[b] Agricultural University of Hebei Province, Baoding 071000,China; University of Hebei Province, Baoding, 071003, China.
hb-ljl@163.com

The incidents of sudden pollution of ammonia nitrogen are happening quite frequently recently. To study the effective removal of ammonia nitrogen, using zeolite powder as an absorbent, the simulation of ammonia-nitrogen wastewater is studied systematically through investigated experiments. The effects of absorption time, ambient temperature, the original ammonia nitrogen concentration, the amount of zeolite and other factors are discussed. Results of experiments show that the absorption performance of zeolite is good and has quick absorption rates. After 10 minutes it basically reaches the absorption equilibrium. At 10~60 ℃, the range effect of temperature on absorption efficiency is rather small. Zeolite absorption of ammonia nitrogen is fitting for Freundlich absorption isotherm equation very well. R2 is 0.9977; parameter n is 1.82, which shows that the zeolite absorption of ammonia nitrogen is better, as the preferential absorbent.

1. Introduction

With increasingly serious water pollution, sudden ammonia nitrogen contamination of waterworks and wastewater treatment plants is breaking out quite frequently. The ammonia nitrogen pollution can be attributed to many reasons, such as extensive use of fertilizers in agricultural production, and highly concentrated urban population. The effects of ammonia nitrogen on microorganisms in water can be broken down into nitrite nitrogen, and then be further purified. However, high nitrite nitrogen in water will combine with protein and then produce carcinogenic nitrosamines which greatly threaten human health. Hence, a study of the effective method to remove ammonia nitrogen is of great significance. Methods of removing ammonia nitrogen in the traditional treatment process include physical method, chemical method and biological method, all of which are difficult to remove the solubility of ammonia nitrogen effectively. Common wastewater biological denitrification process requires strict control of water temperature, PH and other factors, which makes it even harder to achieve compliance with the effluent ammonia nitrogen and is of low efficiency with high operating costs. At present, the physicochemical denitrification by zeolite materials is under the spotlight. Removal of ammonia nitrogen by zeolite includes both ion exchange and absorption. The author used the absorption of zeolite powder materials and analyzed the effects of temperature, the original shock time and solution concentration on zeolite absorption of ammonia nitrogen. In the meantime, change the amount of zeolite and study its isothermal absorption curve. Experiments of ammonia nitrogen by zeolite powder in the course of processing the application provide a theoretical basis.

2. Materials and methods

2.1 Main materials and equipment

2.1.1 Experimental materials

Experiments using zeolite powder, which is made of finely ground natural zeolite stone, were conducted in Hebei Chengde. Its color is cream yellow. It contains mainly Na, Al, Si, O, H and other elements and is insoluble in water. It was soaked for 24h in distilled water before the experiment, and was heated at 105 ℃ for 24h to rule out other influences. Ammonia solution of the water was used for manual configuration, which is made of ammonium chloride solved in distilled water at 100℃. Detection of ammonia reagent for detecting instruments supporting pharmaceutical products is supplied directly by manufacturers.

Please cite this article as: Wang J.Q., Jin W.B., Guo H., Wang X.Y., Liu J.L., 2015, Experimental study on ammonia nitrogen adsorption performance of zeolite powder, Chemical Engineering Transactions, 46, 79-84 DOI:10.3303/CET1546014

2.1.2 Experimental equipment

The main experimental equipment are: XZ-0142 multipara meter water quality analyzer manufactured by Shanghai Electronic Technology Limited Company; SHA-B oscillator and electronic balance manufactured by Jiangsu enterprises in China, etc.

2.2 Experimental methods

2.2.1 Study on the effect of vibration time on the adsorption capacity

Take 44mg/L of ammonia nitrogen concentration solution in 100ml, and divide into six bottles of 250ml Erlenmeyer flasks, and put all in the 200mg of zeolite powder. Place it in the constant temperature oscillator to oscillate for 10min, 20min, 30min, 40min, 50min, and 60min. Take supernatant fluid after settling for a while, and test the static absorption of ammonia nitrogen concentration in the solution afterwards.

2.2.2 Study on the influence of reaction temperature on the absorption capacity

Take six Erlenmeyer flasks; the ammonia solution concentration, the amount of usage and the amount of zeolite are as above. Put them in constant temperature oscillators at 10℃, 20℃, 30℃, 40℃, 50℃, 60℃ respectively for 30min. After settling, determine the ammonia nitrogen concentration of the supernatant.

2.2.3 Study on the influence of initial concentration of solution on absorption capacity

Put high/low two sets concentration of ammonia solution into six bottles of 250ml Erlenmeyer flasks. To all add 200mg of zeolite powder. Place it in the oscillator and shake for 30min. After settling, determine the ammonia nitrogen concentration of the supernatant.

2.2.4 Dosing quantity of zeolite and isothermal absorption

Take 21.43mg/L of ammonia solution per 100ml into seven bottles of 250ml Erlenmeyer flasks. Add 0, 30mg, 50mg, 100mg, 200mg, 300mg, 400mg of zeolite powder respectively. Shake them in oscillator at 20℃ for 30min, then after settling, determine the ammonia nitrogen concentration of the supernatant. Use the following equation to calculate the absorption of ammonia nitrogen balance: $q=V(C0-C)/M$ (1) In the equation, q stands for the equilibrium absorption of ammonia nitrogen; V stands for the initial volume of the solution; C0 stands for the original ammonia nitrogen concentration in the solution; C stands for the ammonia nitrogen concentration in the reaction solution and M is the mass of zeolite powder.

2.2.5 Determination of ammonia nitrogen

Ammonia nitrogen content was measured by the multipara meter water quality analyzers. Principle is the same with Nessler's reagent spectrophotometry.

3. Results and discussion

3.1 Effect of vibration time

Under different oscillation times, zeolite absorption of ammonia nitrogen is shown in Figure 1.

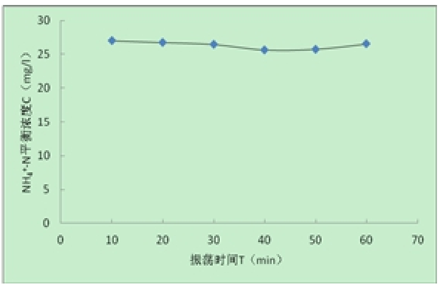

Figure.1 NH4+-N adsorption under different oscillation time

A figure 1 show that during different oscillation times, absorption of ammonia nitrogen concentration in the solution is almost the same, which indicates that time has little effect on the absorption of ammonia nitrogen. Absorption of ammonia nitrogen by zeolite powder can be completed quickly within 10min.

3.2 Effect of reaction temperature

Under different temperatures, the ***equilibrium*** absorption of ammonia nitrogen by zeolite powder is shown in Figure 2.

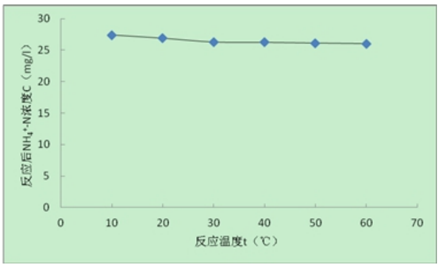

Figure.2 NH4+-N adsorption in different temperatures

As we can see from Table 2, in the range of 10~60℃, as the temperature rises, the ammonia nitrogen concentration slightly decreases after reaction. The highest reduction is 1.34mg/L. In different temperature, ammonia nitrogen concentration after reaction is less than 5%, so the difference is very small. It shows that the effect of temperature on zeolite absorption of ammonia nitrogen is very small. At 10℃, the amount of zeolite absorption of ammonia nitrogen is almost equal to other groups of absorption capacity under high temperature conditions, which suggests that the performance of zeolite absorption of ammonia nitrogen in low temperature is still good. It also proposes the possibility to solve the problem, where conventional denitrification process requires high temperature and outflow in winter when it may be difficult to meet the ideal conditions.

3.3 Influence of initial concentration

In order to determine the different initial concentration of ammonia nitrogen of zeolite absorption, take two groups of high and low concentration gradient to study at the same time. Results are shown in Figures 3 and 4.

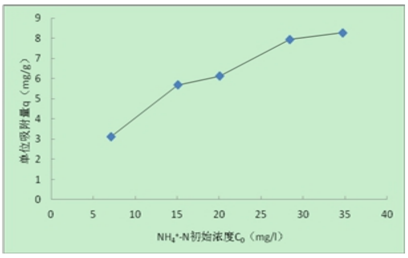

Figure 3: Adsorption of NH4+-N under different initial concentrations (1)

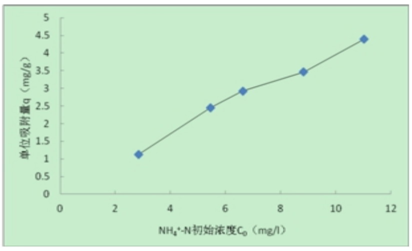

Figure 4: Adsorption of NH4+-N under different initial concentrations (2)

As the tables show, in high concentration and low concentration solution, the zeolite absorption of ammonia nitrogen increases along with the initial concentration of ammonia nitrogen. This is related to the absorption force. The higher the concentration, the greater the absorption force, thus, the greater the absorption capacity.

3.4 Effect of zeolite powder dosing quantity and absorption isotherms

The results of the same initial concentration of ammonia nitrogen but with different zeolite powder dosing amount in the equilibrium concentration of ammonia nitrogen is shown in Figure 5, the absorption capacity in Figure 6.

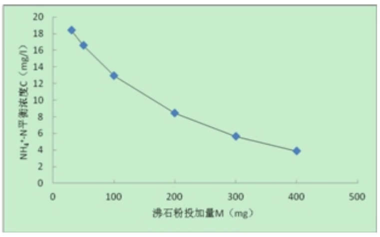

Figure 5: NH4+-N concentration after adsorption with different zeolite powder dosage

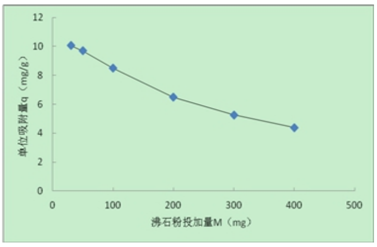

Figure 6: different adsorption of zeolite powder dosage NH4+-N

As we can see from Figure 5, for the same initial concentration of ammonia nitrogen solutions, zeolite absorption of ammonia nitrogen decreases with the increase of dosage. This is because before reaching the saturation concentration, in the same amount and concentration of ammonia nitrogen solutions, the more of zeolite powder dosage, the more units of zeolite absorption potentially decreases. Freundlich and Langmuir absorption isotherms are empirical equations that describe the surface of the water absorption on the solid absorbent. Freundlich adsorption isotherm is: q=KC1/n (2) In the equation, K is Freundlich absorption coefficient; N is Freundlich constants. Usually we use it in a double logarithmic form to determine the accuracy of models. In the form of logarithms, the results are as follows: log q=1/n log C+log K (3) Take log C as X factor and log q as Y factor to make a plane coordinate system. Observe it, to determine whether it is a straight line or not. 1/n is the slope of the straight line and log K its ordinate at the origin. Langmuir adsorption isotherm is: q=qmKLC/ (1+KLC) (4) Which is a straight line in (C/q)~C coordinate. Isotherm converts into: C/q=C/qm+1/KLqm (5) Zeolite absorption of ammonia nitrogen on model fitting curve is shown in Figures 7 and 8. Fitting parameters are shown in Table 1.

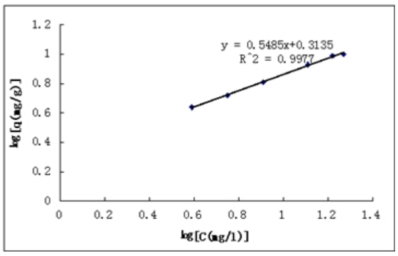

Figure 7: Freundlich adsorption isotherm equation of linear regression

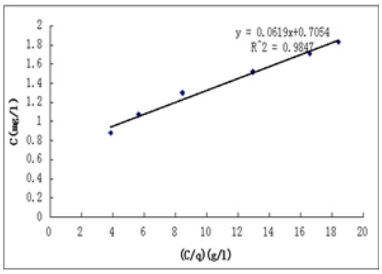

Figure 8: Langmuir adsorption isotherm equation of linea regression

Table 1: Parameters for ammonia nitrogen of zeolite adsorption isotherm

Freundlich			Langmuir		
n	log K	R2	qm	KL	R2
1.82	0.3135	0.9977	16.16	0.088	0.9847

As we can see from Table 6, under single-factorcontrolled conditions, Freundlich and Langmuir absorption isotherm equation could be more fitting for the absorption of ammonia nitrogen by zeolite powder. The correlation coefficient R2 is 0.9977 and 0.9847 respectively. Freundlich isotherm could be more fitting. And at the same time, the uniformed surface monolayer absorption, which Langmuir model requires, is difficult to achieve. So Freundlich absorption isotherm model can better describe the absorption process. Freundlich absorption isotherm parameter N in the model is 1.82, which means zeolite absorption is a preferential absorption of ammonia nitrogen. It is considered that 1/n in between 0.1 and 0.5 is easy to absorb, and more than 2 is hard to absorb. 1/n obtained from Freundlich equations is slightly larger than 0.5, which indicates zeolite easily adsorbs the ammonia nitrogen in water.

4. Conclusions

(1) Absorption of ammonia nitrogen can be completed in a short period of time. Oscillation and the reaction temperature have little impact on the results. The reaction conditions are stable and are conducive to practical application.
(2) Freundlich absorption isotherm equation could be more fitting for the absorption by zeolite powder of ammonia nitrogen in water and R2 reaches 0.9977.
(3) N obtained from Freundlich equations is greater than 1. The absorption of ammonia nitrogen by zeolite powder has good performance and can be preferential for absorption.
(4) This article focuses on the effect of zeolite absorption of ammonia nitrogen and its rate. Experiments show that the ammonia nitrogen removal efficiency of the zeolite powder at high and low concentrations is effective. It has a wide range of application as well as having a fast reaction rate. Its temperature limit is weak and has good practicability.

Reference

Davis M.E., Lobo R.F. 1992, Zeolite and molecular sieve synthesis [J]. Chemistry of Materials, 4 (4): 756-768.

Davis M.E., 1991, Zeolites and molecular sieves: not just ordinary catalysts [J]. Industrial and Engineering Chemistry Research, 30 (8): 1675-1683.

Huang H.M., Xiao X.M., Yan B., et al. 2010, Ammonium removal from aqueous solutions by using natural Chinese (Chende) zeolite as adsorbent. Journal of Hazardous Materials, 175 (1-3): 247-252

Jiang X., Zhou X.N., Ding M.Y., et al. 2008, Natural zeolite and modified zeolite to remove low concentration ammonia nitrogen [J]. Environmental Science, 21 (5): 37-42.

Jin G.H., Jiang H., 2005, Discussion on Causes and Countermeasures of ammonia nitrogen pollution of surface water [a]. Association for Science and Technology, Lishui City, Zhejiang Province, Jinhua City, Zhejiang Province Association for Science and Technology, Quzhou City, Zhejiang Province Association for Science and Technology.The Second Session of the central and western Zhejiang Science and Technology Forum anthology volume eighth (Green Book) [C]. Association for Science and Technology, Lishui City, Zhejiang Province, Jinhua City, Zhejiang Province Association for Science and Technology, Quzhou City, Zhejiang Province Association for Science and Technology, 2005: 4.

Jin X.C., He K., Lu S.Y., Hu X.Z., et al. 2008, Effect of 4 kinds of fillers on the adsorption of ammonia nitrogen [J]. Lake Science, 06: 755-760.

Kurosawa K., Egashira K., Tani M., 2013, Relationship of arsenic concentration with ammonium-nitrogen concentration, oxidation reduction potential and pH of groundwater in arsenic-contaminated areas in Asia. Physics and Chemistry of the Earth. 58-60: 85-88.

Lei L.C., Li X.J., Zhang X.W., 2008, Ammonium removal from aqueous solutions using microwave-treated natural Chinese zeolite. Separation and Purification Technology, 58 (3)：359-366

Li Y., Li J.M., Pan T., 2011, Summary of ammonia nitrogen pollution in groundwater and Treatment Technology [J]. Environmental Engineering, S1:100-102+92.

Malekian R., Abedi-Koupai J., Eslamian S.S., et al. 2011, Ion-exchange process for ammonium removal and release using natural Iranian zeolite. Applied Clay Science,51(3): 323-329

Penn C.J., Warren J.G., Smith S., 2010, Maximizing Ammonium Nitrogen Removal from Solution Using Different Zeolites. Journal of Environmental Quality. 39 (4): 1478-1485.

Peng J.L., 2009, Study on New Type of copper - based ion exchange resin treatment of Ammonia - nitrogen wastewater [d]. Central South University.

Saltali K., Sari A., Aydin M., 2007, Removal of ammonium ion from aqueous solution by natural Turkish (Yildizeli) zeolite for environmental quality. Journal of Hazardous Materials, 141 (1): 258-263.

Sun Z.M., Qu X.S., Wang G.F., Zheng S.L., Frost R.L., 2015, Removal characteristics of ammonium nitrogen from wastewater by modified Ca-bentonites. Applied Clay Science, 107: 46-51.

Tang D.Y., Zheng Z., Guo Z.B., Lin Z.R., Dong C., Wang X.L., 2011, Modification of Low Concentration Ammonia nitrogen wastewater by zeolite adsorption and desorption study [J]. Journal of Environmental Engineering, 02: 293-296.

Tsitsishvili G.V., Andronikashvili T.G., 1992, Natural zeolites [M]. Chichester, England: Ellis Horwood Limited,

Wang Xingnan. 2013 Study on Treatment of High ammonia nitrogen wastewater by immobilized microorganism [d]. Lanzhou Jiaotong University

Yang W., Tian J.Y., Li G.B., 2008, Bioactive filter factors influencing ammonia nitrogen removal from drinking water [J]. Journal of Chemical Engineering, 09: 2316-2321.

Yang Y.F., 2012, Research of Sudden ammonia nitrogen and organic pollution of water treatment technology [d]. South China University of Technology,

You S.H., Tong X.W., Zhu Y.N., 2010, Natural zeolite adsorption of ammonia nitrogen and Its Influencing Factors [J]. Water resources protection, 01: 70-74.

Zhang J., Li D.P., Tao Y., Wang X.M., He X.H., Gao P., 2010, Pollution of ammonia nitrogen in Jinjiang and biological transformation characteristics [J]. Journal of Sichuan University (Natural Science Edition), 03: 619-625.

Zhang L.S., Wang Y.Y., Meng F.S., Liu T.L., Ma G.W., Zhou Y.X., LiuY., 2013, Study on Characteristics of Ammonia - nitrogen pollution in Songhuajiang River Basin [J]. Environmental Science and Technology, 10:43-48+77.

Zhang T., Li Q.C., Ding L.L., Ren H.Q., 2011, Modeling assessment for ammonium nitrogen recovery from wastewater by chemical precipitation. Journal of Environmental Sciences.23 (6): 881-890.

Zhang X.Y., Wu Z.C., Wang Z.W., and so on. 2010, Natural zeolite powder on Adsorption Mechanism of solution NH4+ [J].China Environmental Science, 30 (5): 609-614

Zhang Y., 2001, Ammonia - nitrogen pollution in rivers, Hubei Province, and [J]. Environmental Science and Technology, S2: 30-31.