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STUDY OF THE PERFORMANCE OF BATCH BIOLOGICAL DENITRIFICATION USING LIGNOCELLULOSIC MATERIALS AS A CARBON SOURCE IN THE TREATMENT OF LOW C/N WATER

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Abstract

Nitrate pollution of groundwater and drinking water has become an increasingly serious problem across the world. In Algeria, pollution of groundwater by nitrates has become alarming. Excess nitrates in drinking water can cause childhood methemoglobinemia (blue baby syndrome) and the reduction of nitrate to nitrites in saliva can induce the formation of nitrosamines, which are known carcinogens. Among the different technologies available for nitrate removal involving ion exchange, reverse osmosis, electrodialysis, heterotrophic denitrification seem to be the most promising processes.In this process, the bacteria uses the nitrate for respiration, converting it to nitrogen gas through a series of reactions:

$$NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2$$

In the present study, in order to remove nitrates from simulated groundwater contaminated with nitrates, a natural organic substance; acorn cup powder was used as a carbon source and as a biofilm support for batch denitrification. The effect of various experimental conditions such as initial nitrate concentration, amount of carbon source and pH was studied.Batch tests were carried out in a 1L Erlenmeyer flask, 100 mL of inoculum was added to 900 mL of synthetic groundwater prepared with KNO_3, mineral salts were added. The results obtained show that the system achieved high denitrification, nitrate removal was above 98% after 24 hours of incubation when the pH was 7, NO_3^- = 70 mg/L and 8 g/L concentration powder from acorn cups. The NO_2^- level in the effluent was less than 0.07 mg/L. Nitrate removal was above 95% after 3 hours of incubation when pH was 7.1, NO_3^- = 70 mg/L and 3 g/L biomass concentration. The NO_2 level in the effluent was less than 0.0030 mg/L. Our results suggest that it is possible to use reed powder in denitrification with additional treatment for drinking water.

Keywords

Biological denitrification, acorn cups, Nitrate, Water treatment

I. Introduction

Due to agricultural runoff and treated wastewater discharged to receiving waters, nitrate pollution of groundwater and drinking water has become an increasingly serious issue throughout the world since the 1970s1 . Excess nitrates in drinking water can cause infantile methemoglobinemia (blue baby syndrome) and reduction of nitrate into nitrites in saliva might induce the formation of nitrosamines known as carcinogens. The nitrate levels of lower than 10 mg NO3 — N /L are regulated by the US Environmental Protection Agency and 50 mg NO3 - /L by World Health Organization and European Economic Community to prevent the risks to human health4 .

Among the various technologies available for nitrate removal involving ion-exchange, reverse osmosis and electrodialysis5 , heterotrophic denitrification seems to be the most promising process6 . It proves more economical, practical on a large scale and presents a permanent solution by ultimately reducing nitrate to nitrogen gas which returns reactive nitrogen to the atmosphere and maintains the balance of the global nitrogen budget. Recently, solid phase denitrification using solid substances involving natural organic substances such as woods chips, wheat straws, cottons, maize cobs, reeds etc. and synthetic biodegradable polymers serving as carbon source for denitrification and biofilm carriers, has proved to be promising alternative to remove nitrate from water and wastewater. In this process, heterotrophic bacteria use organic carbon for both growth and as an electron source. The efficiency of the process depends on an adequate supply of organic carbon that can act as an electron donor for the reduction of nitrate to nitrite and finally to nitrogen gas. The bacteria uses nitrate for respiration, converting it to nitrogen gas by a sequence of reactions:

$$NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2\uparrow$$

In the present study, in order to remove nitrate from simulated nitrate-contaminated groundwater, a natural organic substances reed powder and acorn cup powders were used as carbon source and biofilm carrier for denitrification in batch. The effect of varied experimental conditions as initial nitrate concentration, carbon amount change and pH was investigated

2. Material and Methods

Batch experiments were performed to investigate the water denitrification efficiency using reed powder and acorn cup powders as carbon source and biofilm carrier; they were washed with distilled water, dried under laboratory conditions, crushed, reduced, passed through 1.25 mm sieve and then preserved at 280C. The same collected material was used in all experiments .

Batch assays were conducted in 1L Erlenmeyer flask (fig. 1). The experiments were conducted in total darkness; the systems were covered with aluminium paper to prevent light penetration and under anoxic conditions (the headspace gas contained O2 initially, but it was rapidly consumed by bacteria). The inoculum used was a mixture of 50 mL of adapted activated sludge from a domestic wastewater treatment plant in Boumerdes, Algeria and 50 ml without acclimation to nitrate.

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(Inoculation with adapted activated sludge to the substrate and operating conditions is a serious gain of time). 100 mL of the inoculum was added into a 900 mL of synthetic groundwater prepared with KNO3 (50 -130 mg/L NO3 -), the mineral salts media contained the following final concentration in mg/L: CaCl2.2H2O (1.32), FeCl3.6H2O(0.12), K2HPO4(5), MgSO4.7H2O (2), (NH4)6 Mo7O24.4H2O(0.01), ZnSO4.7H2O(0.1) and trace elements composed of CoCl2.2H2O(0.01), CuSO4.5H2O (0.02), NiSO4.6H2O(0.01), H3BO3(0.005), MnCl2.4H2O (0.023)12,20,24 . The reactors were mixed using a magnetic stirrer at 250 rpm in all phases except for the settling and decanting phases

At the end of each experiment, the supernatant was collected using a syringe for nitrate, nitrite and pH analysis according to standard methods for the examination of water and wastewater. The volume of 80 ml of sample is taken for analysis and an equal volume of the nitrate solution is added.

Adapted activated sludge was obtained by cultivating the mixture of 100 ml of activated sludge,separately 1 g of reed powder or acorn cup powders and 900 ml of nitrate solution at 50 mg/L of NO3 – are added with mineral salts. The reactors were inoculated and maintained in batch mode for 21 days.

For the study of the pH influence on biological denitrification, synthetic water was adjusted to the desired pH by changing the ratio of phosphate salts (K2HPO4 and NaH2PO4.H2O). The NO3 - removal efficiency during the experimental period was calculated based on the following equation:

R (%)= (Cin – Cen) *100 /Cin

where Cin (mg/L) and Cen (mg/L) were the concentrations of NO3 - in the flasks at the beginning and end of the study respectively.

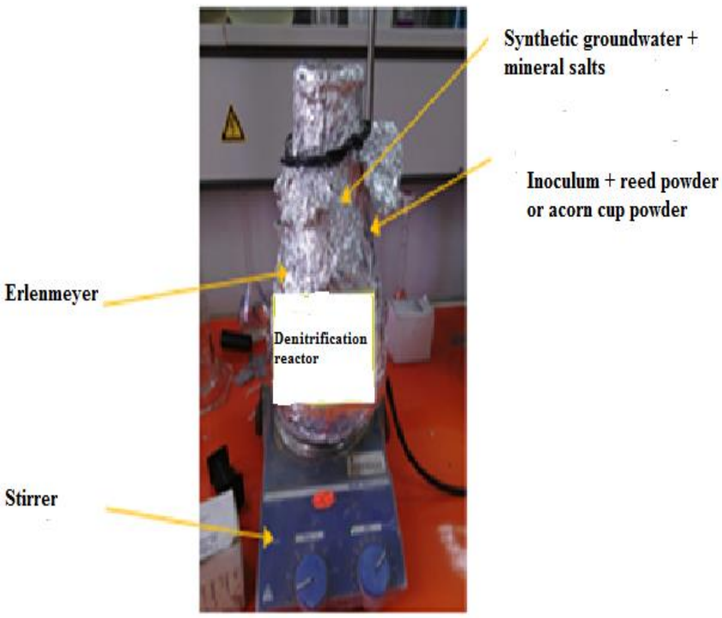

Figure 1: Biological denitrification in batch reactor

3.Results and Discussion

3.1Chemical compounds of the sawdust:

The main chemical compounds were determined for: humidity (Hu), total ash (TA) as well as some mineral elements analyzed by atomic absorption spectrometry (Zn, Cu, Fe, Mn, Cd, Pb). The chemical composition of reed powder and acorn cup powder is given in table1. The moisture content indicates the stability of the products against the risks of deterioration during storage. According to the results found (table 1), reed powder and acorn cup powder have respectively 1.74 and 3.45 ash content, therefore they are very rich in organic matter such as carbon which is a nutrient used as source energy for growth and development of denitrifying bacteria. They are rich in nutrients such as Zn2 + and Fe + 2 which are constituents of certain enzymes necessary for the growth and development of denitrifying bacteria.

Table 1 : Chemical composition of reed powder and acorn cup powder

Parameters	Value		Référence
	Reed powder	Acorn cup powder	
Hu (%)	6.14	6.24	NF V 03-903
TA (%)	1.74	3.45	NF V 05-113
Zn (µg/g)	70	86.3	NF V 05-113
Cu (µg/g)	25.46	8.83	NF V 05-113
Fe (µg/g)	213.4	65.85	NF V 05-113
Mn (µg/g)	/	145.9	NF V 05-113
Cd(µg/g)	0.303	0.432	NF V 05-113
Pb (µg/g)	0.435	2.903	NF V 05-113

3.2.Studies of parameters influencing denitrification with Reed powder

3.1.1.Effect of initial nitrate concentration

The advancement of the sludge age significantly improves the denitrification efficiency, so the sludge added to the reactor is composed of 50 ml of already adapted bacteria and 50 ml of young bacteria.

Nitrate removals are presented as a function of initial nitrate concentration at the end of the 2 h d' incubation (fig. 2). The data indicated that when initial nitrate concentration increased to 130 mg/L, NO3 - removal efficiency increased to approximately 93%. This can be explained by the fact that when the initial amount of nitrate increases, the amount of enzyme nitrate reductase increases and thus denitrification improves.

Figure 3 showed also that the accumulation of NO2 - as intermediate product was low (less than 0.009 mg/L) with 50 to 130 mg/L NO3 - . The test resulted in a slight fluctuation of pH from 6.95 to 7.35 (figure 4). In the view of these results, denitrification was slightly influenced by initial nitrate concentration and we noted a good nitrogen removal performance over a wide range of nitrate concentrations.

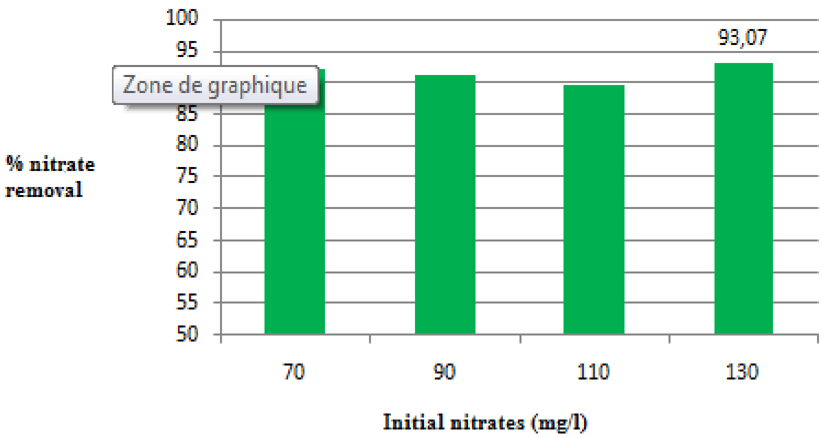

Figure 2: Effect of initial nitrate concentration on denitrification (Conditions: pH=7.0; biomass concentration=1g/L and Time = 2 hours)

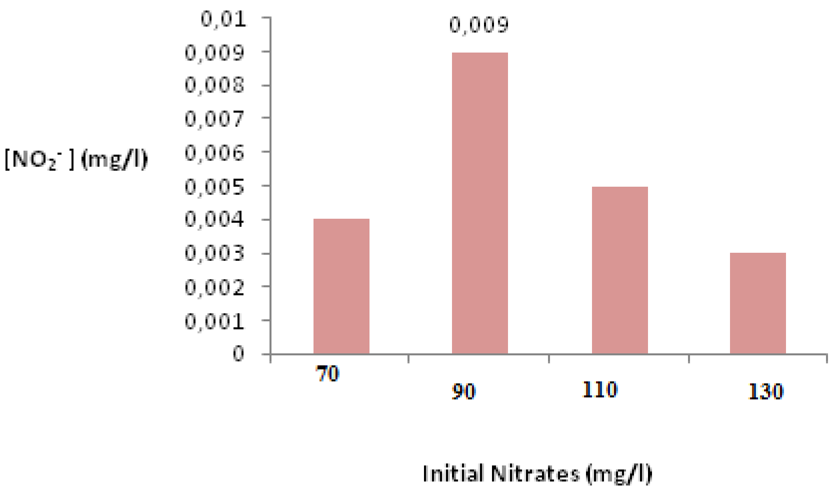

Figure 3: NO2 - levels in the effluent (**Conditions: pH=7.0; biomass concentration=1g/L and Time = 2 hours)**

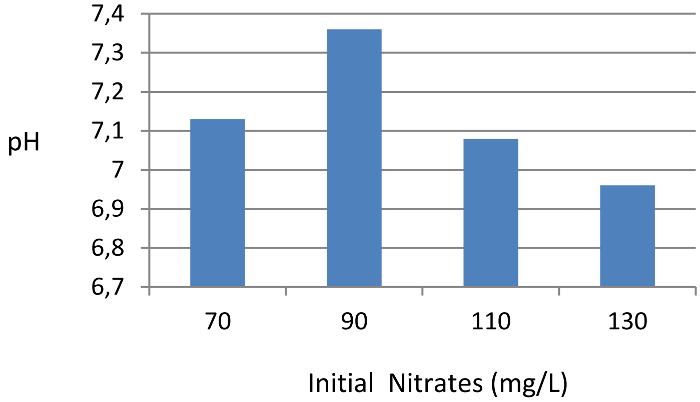

Figure 4: The pH in the effluent (**Conditions: pH=7.0; biomass concentration=1g/L and Time = 2 hours)**

3.1.2.Effect of biomass concentration

The carbon to total nitrogen ratio is a measure of the electron donor to acceptor ratio in biological denitrification. To determine the influence of carbon amount on nitrate removal, the biomass concentration was varied from 1 to 8 g /L. At the end of the 2 h incubation, the results (fig. 5) showed that the denitrification efficiency increased with the increasing carbon source amount until reaching complete denitrification with biomass concentration of 6 g/L. As described previously by Ovez, increasing carbon source increased the adaptation surface area for the denitrifying population. A high potential denitrification velocity would thereby be the result of more metabolically active bacteria in the denitrification medium. In biological denitrification, nitrite production is known to be one of the main limitations because it inhibits bacterial growth and is retained in the water.

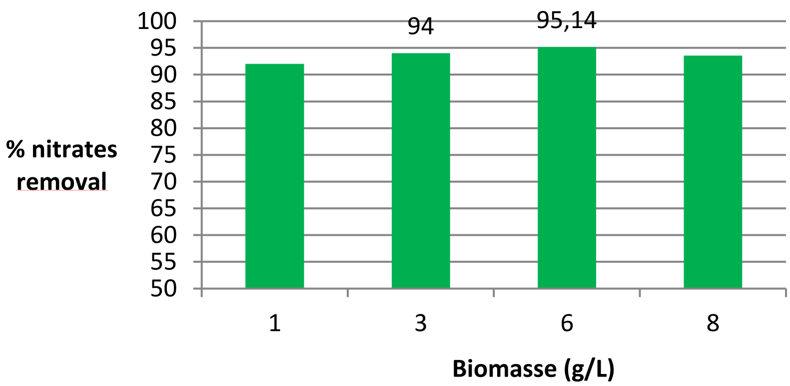

Figure 5: Effect of biomass concentration on the nitrate removal (Conditions: [NO3 -]= 70 mg/L; pH= 7.1

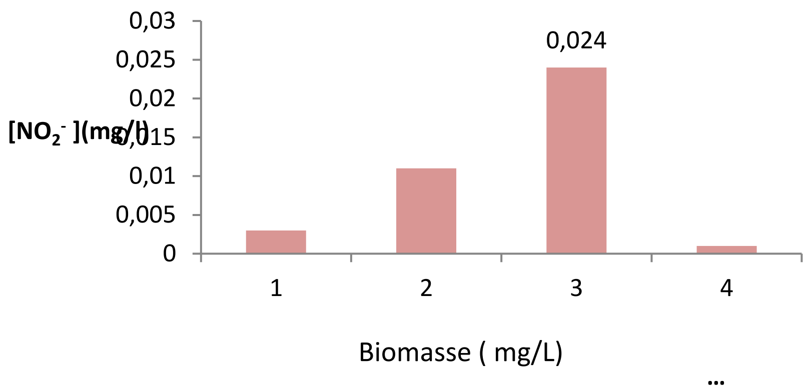

Figure 6: NO2 - levels in the effluent (Conditions: [NO3 -]= 70 mg/L; pH= 7.1)

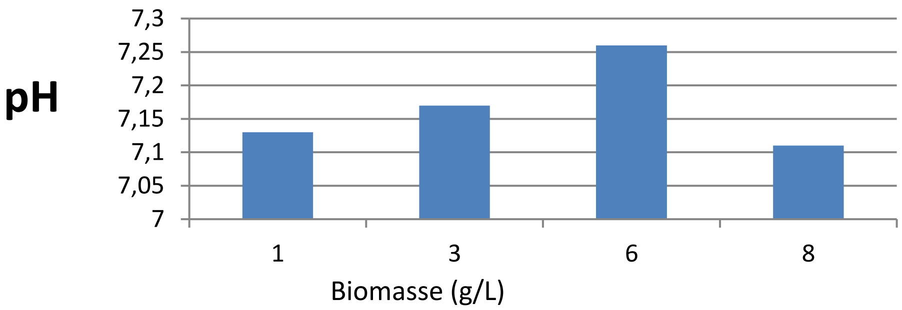

Figure 7 : The pH in the effluent (Conditions: [NO3 -] = 70 mg/L; pH= 7.1 **and Time 2 hours**)

3.1.3.Effect of initial pH on denitrification

The pH has an important influence on the control of biological denitrification. Denitrification can take place in the pH range 4– 11, but most of the bacteria are slightly basophilic with a pH optimum range of 7.5–8.5 33. Denitrification was tested according to initial pH. The different initial pH values were studied in the reactors maintained at a constant C/N ratio (1 g sawdust, 70 mg/L NO3 -). This was done by adding quantities of KH2PO4 to vary the acidity and quantities of Na2HPO4 for the basicity. As seen from figure 8, the pH values investigated resulted in small amounts of variations in the performance of the denitrification reaction; nitrate removal was greater than 91% at the end of the 2 h incubation. The optimal pH is 7.13. However, in the literature, the optimal of pH was found by Yang and Bingfallen34 equal to 7.1. Foglar and Vukovié35 found 7.4 and Lykiardopol et al36 found around 7 to 7.5. The concentration of nitrite was detected at less than 0.45 mg l -1 in the effluent (figure 13)

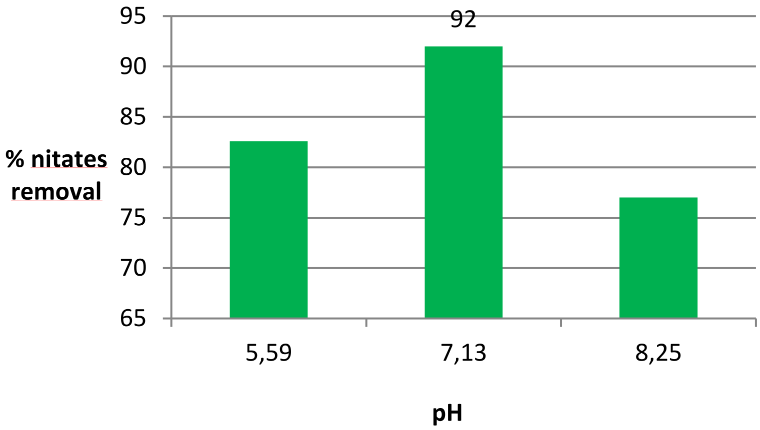

Figure 8: Nitrate removal for different values of pH (Conditions: [NO3 -]= 70 mg/L , biomass concentration=1g /L)

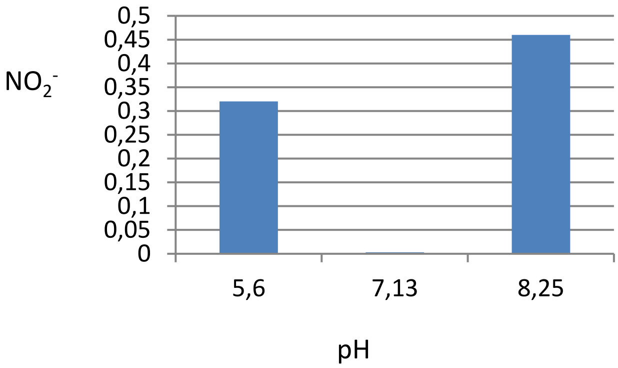

Figure 9: NO2 - levels in the effluent (Conditions: [NO3 -] = 70 mg/L, biomass concentration=1g/L **and time =2 hours**)

3.3. Studies of parameters influencing denitrification with Acorn cup powder

3.3.1 Effect of initial nitrate concentration

Nitrate removals are presented as a function of initial nitrate concentration at the end of the 1day h incubation (fig. 10). The data indicated that when initial nitrate concentration increased from 50 to 110 mg/L, NO3 - removal efficiency increased gradually to approximately 93%. This can be explained by the fact that when the initial amount of nitrate increases, the amount of enzyme nitrate reductase increases and thus denitrification improves. Then, denitrification efficiency decreases slightly for higher initial nitrate concentration 130m/l because of the slight reduction in the C/N of cultures.

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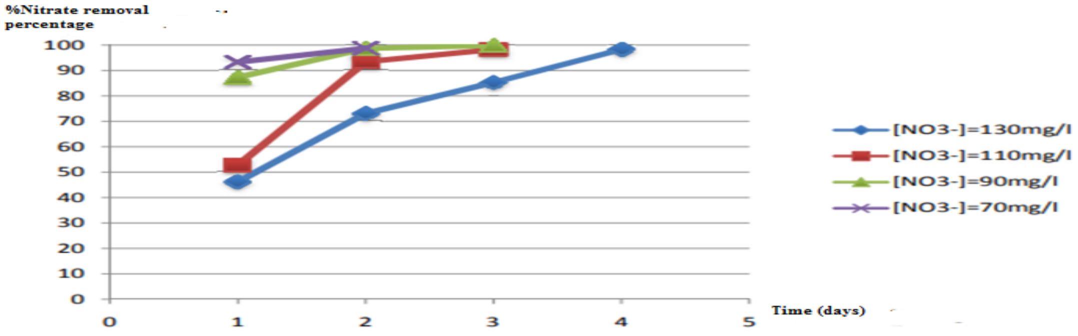

Figure 10: Effect of initial nitrate concentration on denitrification**, pH = 7**

Conditions: pH=7.0; biomass concentration=1g/L)

3.2.2.Effect of biomass concentration

To determine the influence of carbon amount on nitrate removal, the biomass concentration was varied from 2 to 12 g /L. At the end of the 1day incubation, the results (fig. 11) showed that the denitrification efficiency increased with the increasing carbon source amount until reaching complete denitrification with biomass concentration of 8 g/L (97%). 64

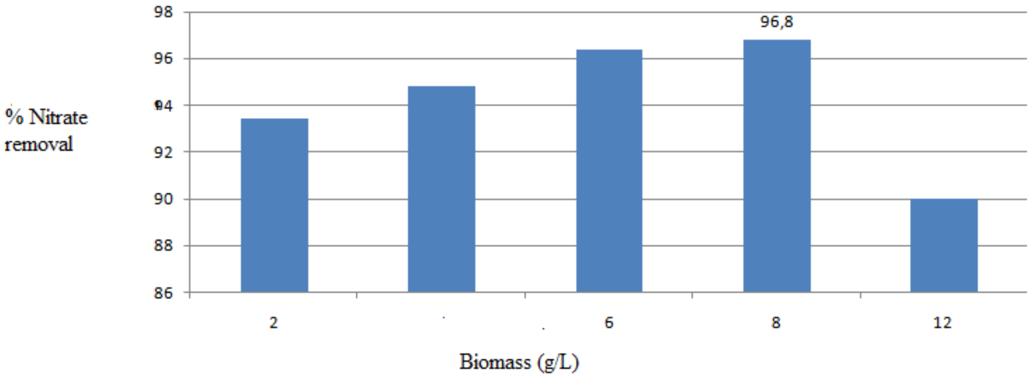

Figure 11: Effect of biomass concentration on the nitrate removal (Conditions: **[NO3-] = 50 mg/L, pH = 7 and time = 1 days**

3.3.3. Effect of initial pH on denitrification

Denitrification was tested according to initial pH. The different initial pH values (4.5, 7 and 11.5) were studied in the reactors maintained at a constant C/N ratio (1 g sawdust, 50 mg/L NO3 - As seen from figure 12, the pH values investigated resulted in small amounts of variations in the performance of the denitrification reaction; nitrate removal was greater than 98% at the end of the 1 day incubation. The optimal pH is 7.

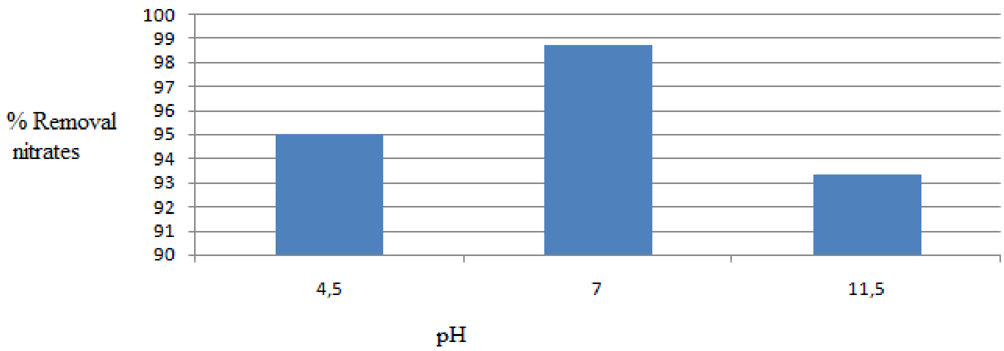

Figure 12: Nitrate removal for different values of pH (**[NO3-] = 50 mg / L, time 1 days and biomassc=1g/L**

4.Conclusion

The results obtained from batch denitrification experiments using reed powder as carbon source and biofilm support show that the system achieved high denitrification, nitrate removal was above 92% after 2 hours of incubation when the pH was 7, NO3- = 70 mg/L and 1g/L biomass concentration. The NO2-level in the effluent was less than 0.030 mg/L. The results also showed a significant effect of C/N ratio on process performance.

For the batch denitrification experiments using acorn cup powder as carbon source and biofilm support, the results obtained show that the system achieved high denitrification, nitrate removal was greater than 98.71% after 1 day of incubation when the pH was 7, NO3- = 50 mg/L and 1g/L biomass concentration. The NO2-level in the effluent was less than 0.030 mg/L.

The results also showed a significant effect of C/N ratio on process performance. Our results suggest that it is possible to use these natural organic substances which are abundant in our country (Algeria) with relatively low price, efficiency and less harmful effects as a carbon source in denitrification with additional treatment for drinking water. However, for industrial application, better parameter optimization is required.

References

1. Gibert O., Pomierny S., Rowe I. and Kalin R.M., Selection of organic substrates as potential reactive materials for use in a denitrification permeable reactive barrier (PRB), Bioresour. Technol., 99(16), 7587–7596 (2008)

2. Matiju V., Cizinska S., Krejci J. and Janoch T., Biological water denitrification - a review, Enzyme Microb. Technol., 14(3), 170– 183 (1992)

3. Nolan B.T., Ruddy B.C., Hitt K.J. and Helsel D.R., A national look at nitrate contamination of ground water, Water Cond. Purif., 39(12), 76-79 (1998)

4. Tsai H.H., Ravindran V., Williams M.D. and Pirbazari M., Forecasting the performance of membrane bioreactor process for groundwater denitrification, J. Environ. Eng. Sci., 3(6), 507-521 (2004)

5. Aslan S. and Turkman A., Simultaneous biological removal of endosulfan (alpha plus beta) and nitrates from drinking waters using wheat straw as substrate, Environ. Int., 30(4), 449–455 (2004)

6. Park J.B.K., Craggs R.J. and Sukias J.P.S., Removal of nitrate and phosphorus from hydroponic wastewater using a hybrid denitrification filter (HDF), Bioresourc. Technol., 100(13), 3175– 3179 (2009)

7. Mike S.M.J., The microbial nitrogen cycle, Environ. Microbiol., 10(11), 2903-2909 (2008)

8. Ovez B., Mergaert J. and Saglam M., Biological denitrification in drinking water treatment using the seaweed Gracilaria verrucosa as carbon source and biofilm carrier, Water Environ. Res., 78(4), 430–434 (2006a)

9. Schipper L.A., Robertson W.D., Gold A.J., Jaynes D.B. and Cameron S.C., Denitrifying bioreactors-an approach for reducingnitrate loads to receiving waters, Ecol. Eng., 36(11), 1532–1543 (2010b)

10. Lu H., Chandran K. and Stensel D., Microbial ecology of denitrification in biological wastewater treatment, Water Res., 64, 237-254 (2014)

11. Chu L. and Wang J., Denitrification performance and biofilm characteristics using biodegradable polymers PCL as carriers and carbon source, Chemosphere, 91(9), 1310–1316 (2013)

12. Ovez B., Batch biological denitrification using Arundo donax, Glycyrrhiza glabra and Gracilaria verrucosa as carbon source, Process Biochemistry, 41(6), 1289–1295 (2006)

13. Xu Z.X., Shao L., Yin H.L., Chu H.Q. and Yao Y.J., Biological denitrification using corncobs as a carbon source and biofilm carrier, Water Environ. Res., 81(3), 242–247 (2009)

14. Robertson W.D., Nitrate removal rates in wood chip media of varying age, Ecol. Eng., 36(11), 1581–1587 (2010)

15. Cameron S.G. and Schipper L.A., Hydraulic properties, hydraulic efficiency and nitrate removal of organic carbon media for use in denitrification beds, Ecol. Eng., 41, 1–7 (2012) 16. Hiraishi A.

and Khan S.T., Application of polyhydroxyalkanoates for denitrification in water and wastewater treatment, Appl. Microb. Biotechnol., 61(2), 103–109 (2003)

17. Boley A. and Muller W.R., Denitrification with polycaprolactone as solid substrate in a laboratory-scale recirculated aquaculture system, Water Sci. Technol., 52(10−11), 495–502 (2005)

18. Chu L. and Wang J., Denitrification of groundwater using PHBV blends in packed bed reactors and the microbial diversity, Chemosphere, 155(3), 463–470 (2016)

19. Chu L. and Wang J., Nitrogen removal using biodegradable polymers as carbon source and biofilm carriers in a moving bed biofilm reactor, Chemical Engineering Journal, 170(1), 220–225 (2011)

20. Saliling W.J.B., Westerman P.W. and Losordo T.M., Wood chips and wheat straw as alternative biofilter media for denitrification reactors treating aquaculture and other wastewaters with high nitrate concentrations, Aquacul. Eng., 37(3), 222–233 (2007)

21. Lee K. and Rittmann B.E., Effects of pH and precipitation on autohydrogenotrophic denitrification using the hollow-fiber membranebiofilm reactor, Water Res., 37(7), 1551–1556 (2003)

22. Healy M.G., Rodgers M. and Mulqueen J., Denitrification of a nitrate-rich synthetic wastewater using various wood-based media materials, J Environ Sci Health A Tox Hazard Subst Environ Eng., 41(5), 779–788 (2006) 23. Park W., Jang E., Lee M.J., Yu S. and Kim T.H., Combination of ion exchange system and biological reactors for simultaneous removal of ammonia and organics, Journal of Environment Management, 92, 1148–1153 (2011)