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BIOLOGICAL NITRIFICATION-DENITRIFICATION TREATMENT OF AMMONIUM NITROGEN POLLUTED HANOI-GROUNDWATER USING A NOVEL ACRYL-RESIN FIBER FOR BIOMASS ATTACHMENT

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Abstract

This study demonstrates a study research on biological nitrification-denitrification treatment of ammonium nitrogen polluted Hanoi-groundwater using a novel acryl-resin fiber for biomass attachment. The denitrification batch assays with synthetic groundwater and the continuous flow experiments with ammonium contaminated raw groundwater at Phap Van water treatment plant were conducted with three kinds of supplemental carbon sources of glucose, ethanol and methanol. Research results consistently revealed that ethanol could be a best supplemental carbon source for the denitrification treatment of ammonium contaminated groundwater and the use of biological nitrification-denitrification system using a BL material as a biomass attachment medium has demonstrated potential for removal of ammonium from polluted groundwater in Hanoi. The system can play an effective role in providing safe drinking water.

Keywords: Groundwater, ammonium, nitrification, denitrification, biomass carrier, biofill.

1. Introduction

Over the last ten years the population of Hanoi has increased rapidly to more than 3 millions people. In the city there is an intensive exploitation of groundwater for domestic and industrial purposes. The Hanoi water business company is exploiting main well fields with about 150 wells for the 10 water treatment plants with the total production capacities of 400,000 to 450,000 cubic meters per day. Beside that, there are more than 500 industrial production wells with smaller capacity that are exploited by enterprises, research institutes, hospitals, school units ... with approximately 50,000 to 60,000 cubic meters per day exploited in the city. All around city there are more than 100,000 small bore wells (so called UNICEF wells) with total capacity of about 200,000 cubic meters per day. Thus, the total volume of groundwater exploited in Hanoi area is up to 600,000 – 650,000 cubic meters per day. This volume is nearly reaching a limit of groundwater availability for the city. Under such intensive and exhausting exploitation the groundwater resource is facing the serious risk of table depletion and quality deterioration.

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In recent years, ammonium concentration in the groundwater in Hanoi has become an increasing problem for water quality management in the city. The most heavily polluted areas are located mainly in the southern part of the city where nitrogen ammonia concentrations range from 20 to 30 mg/L.

According to the results drawn from different previous studies (Ha C.T., 2004, HWBC, 1997 – 2004), ammonium content in water of the aquifers is changing from trace to 30 mg/L. Most of the areas are polluted by ammonium as Phap Van, Dinh Cong, Ha Dinh, Kim Giang, Tuong Mai, Bach Mai, Bach Khoa, Kim Lien, and Quynh Mai.

Among of them, some areas are lightly polluted as Luong Yen, Yen Phu, Ngo Si Lien, and Don Thuy. Besides, some areas have a sign of pollution as Ngoc Ha, Mai Dich. In general, the strongly polluted areas are mainly located in the south and the center. There is the highest pollution (~ 20 mg/L) in Phap Van, after that in Ha Dinh area (~ 12 mg/L), Tuong Mai (~ 10 mg/L). It is worth to note that in the monitored areas, the nitrate and nitrite content are not high, meeting drinking water standard (Fig. 1.1).

Table 1.1 The Vietnamese and international standard for nitrogen compounds in supply water (Ha C.T. et al, 2004).

Parameter	Unit	EPA Standard	EC standard (80/778/EEC)		WHO Guidelines (1993)	Vietnamese standards		
		MCL	GL	MAC		(1)	(2)	(3)
Ammonium (NH_4^+)	mg/L			0.5	1.5	3 ^a	3 ^a	1.5 ^a
Nitrate (NO_3^-)	mg/L	44.3	25	50	50	6(as N)	22.1	50
Nitrite (NO_2^-)	mg/L	4.4		0.1	3	0	0	3
COD (KMnO_4)	mg/L					2	2	2

(1) 20 TCN 33-85: Construction code issued by Ministry of Construction (1985).

(2) Vietnamese Standard TCVN 5501-91 for drinking water quality (1991).

(3) Vietnamese Standard 1329/2002 - BYT/QD issued by Ministry of Health (2002).

^a Groundwater.

MCL - Maximum contaminant levels.

GL - Guidelines.

MAC - Maximum acceptable concentration.

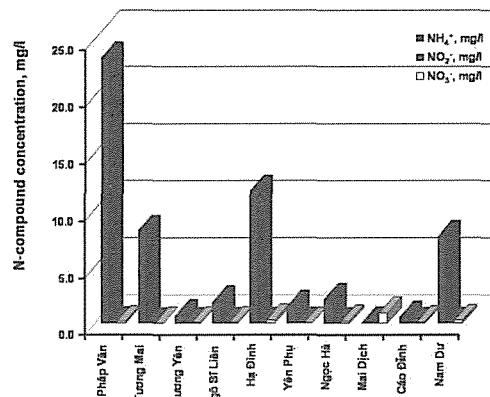


Fig. 1.1. Concentration of nitrogen compounds in main water treatment plants in Hanoi city (Ha C.T. et al., 2004)

Before 2002, in Vietnamese standard, the ammonium content in the supply water was required to be lower than 3 mg/L. Since April 2002, according to the new standard No. 1329/2002 – QD/BYT, the

ammonium content must be under 1.5 mg/L, equal to the World Health Organization (WHO) standard. The standard of the European Union for ammonium is under 0.5 mg/L. (**Table 1.1**).

Treatment technologies for nitrogen ammonium removal from groundwater supply are still unknown not only to Vietnam but also to many other countries in the world, especially at raised ammonium concentration of 10 - 20 mg/L. This is a reason to promote this research. It is needed to find an appropriate method of high efficiency, stable operation, acceptable cost where treated water should meet the Vietnamese water quality standards.

In the previous research, we have investigated a new method for biological nitrification of groundwater contaminated with ammonium using acryl-resin fiber (BL material) as a biomass carrier material. High rate of nitrification efficiencies of 95~99% at ammonium loading rates up to 0.75 g-N/l.d were demonstrated. However, the research was conducted under laboratorial condition with synthetic groundwater and focused on the nitrification step.

The propose of this continuous phase of research was:

- To investigate the suitable carbon source for denitrification process; and
- To evaluate the applicability of a biological nitrification-denitrification process using acryl resin for ammonium removal from polluted groundwater in the southern part of Hanoi.

2. Materials and methods

2.1 Cultivation of denitrifying activated sludge

5 l of activated sludge, donated by the Vietcombank Wastewater Treatment plant, was used as seed sludge and cultivated in 15 l bucket by fill and draw under anoxic conditions. A synthetic substrate (**Table 2.1**) was used for cultivation of denitrifying activated sludge. The growth rate of sludge was estimated using mixed-liquor suspended solids (MLSS).

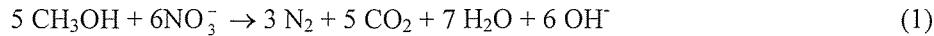
2.2 Batch assay

In order to investigate the denitrification ability of sludge with different carbon sources, methanol, ethanol and glucose were used. The experiment was conducted in 3 denitrification reactors (5L) and the concentration of denitrifying sludge was set at 2500 mg/l. NO_3^- -N concentration in substrate was 1 g/l, methanol, ethanol and glucose were added with concentrations of 1,9; 1,4 and 2,68 g/L, respectively, according to the following equations:

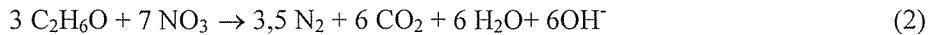
Table 2.1 Composition of the synthetic groundwater (mixed in tap water).

Composition	Concentration (mg/l)	Source
NO_3^- -N	1000	NaNO_3
TOC	1500	$\text{C}_6\text{H}_{12}\text{O}_6$
SO_4^{2-}	2,8	tap water
Fe(II)	0~18	FeCl_2
Ca	25	$\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$
Mg	13	$\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$
Na	35	tap water
K	5,7	tap water
Alkalinity	100~250	NaHCO_3

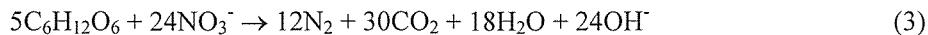
Metanol: CH_3OH



Etanol: $\text{C}_2\text{H}_6\text{O}$



Gluco: $\text{C}_6\text{H}_{12}\text{O}_6$



The liquid of substrate and sludge inside the reactors was mixed by mixer with the rate of 100rpm and the reactors were sealed with rubber covers to maintain an anoxic condition. K_2HPO_4 and KH_2PO_4 were added at 0.14; 0.02 g/L, respectively. And trace nutrients were added in forms of NaCl ; KCl ; $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$; $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ at the concentrations of 0.006; 0.003; 0.005 and 0.004 g/L, respectively.

The batch assay was conducted by fill and draw method under room's temperature. The substrate was made and newly changed every day. After 2 weeks of cultivation, the decrease of NO_3^- -N concentrations was monitored after 1, 3, 6, 12, 24 h.

2.3 Continuous flow assay

In order to evaluate the applicability of a biological nitrification-denitrification process for ammonium removal from polluted groundwater in the southern part of Hanoi, the continuous flow assay was conducted at the Phap Van water treatment plant.

2.3.1 Biomass carrier

In order to immobilize nitrifying activated sludge, an acryl-resin fiber material called Biofill (BL) donated by N. E. T. Co. Ltd. was used. This kind of biomass carrier material is light in weight, inexpensive and durable.

Two strips of the BL material (Fig. 2.1) each with a one-sided surface area of 300x450 mm and a weight of 32.7 g were used as biomass carriers in this study. The BL strips were folded in to 4 layers and set symmetrically on two sides of the nitrification reactor using aluminum frames (see Fig. 2.2). For the denitrification reactor, BL strips were scrolled around the central pipe.

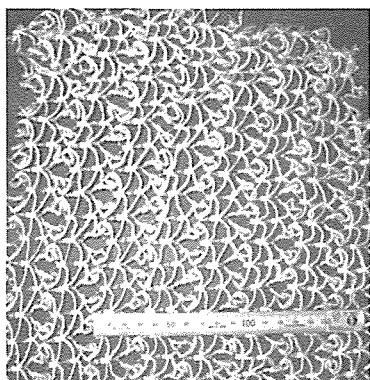


Fig. 2.1 BL material.

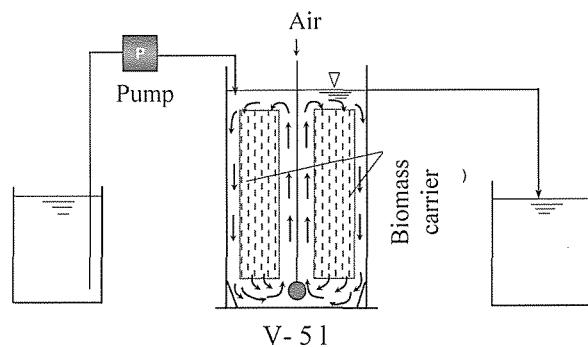


Fig. 2.2 Nitrification reactor.

2.3.2 Nitrification reactor

Fig. 2.2 shows the schematic diagram of the nitrification reactor. The 5-l reactor was made of PVC and the influent was fed by using a variable speed peristaltic pump. BL strips were set symmetrically in the reactor. Aeration at the base of the reactor kept the contents well mixed and oxygenated. Fig. 2.3 shows the pictures of reactor during the start-up period.

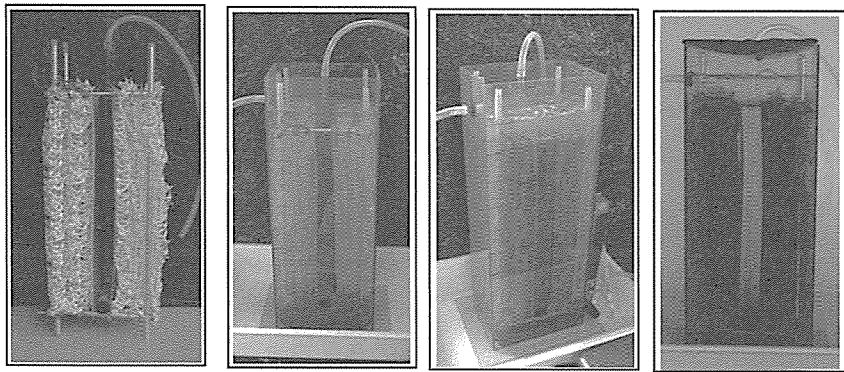


Fig. 2.3 Start-up of the nitrification reactor.

2.3.3 Denitrification reactors

Fig. 2.4 shows the schematic diagram of the nitrification reactors. Each reactor (V-1.85L), made by glass-cylinder columns were fed separately with methanol, ethanol and glucose from the bottom by peristaltic pump and the effluent from the nitrification reactor was pumped to the upper part of the reactor. The mixer inside a central pipe of the reactor kept the contents well mixed. **Fig. 2.5** and **2.6** show the denitrification column and nitrification-denitrification experimental system which were set at Phap Van water treatment plant.

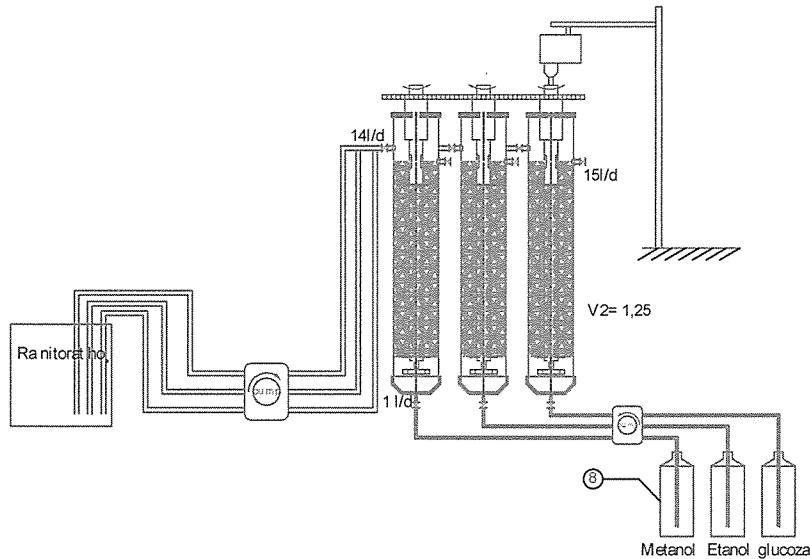


Fig. 2.4 Schematic diagram of denitrification reactors.

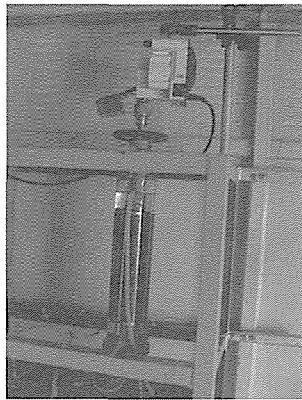


Fig. 2.5 Denitrification column.

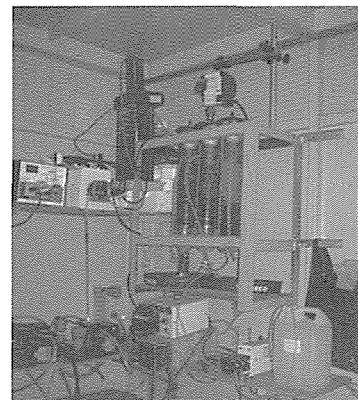


Fig. 2.6 Nitrification-denitrification system.

2.3.4. Operational conditions

The tested groundwater was taken from the outlet of sand-filter of Phap Van water treatment plant. The average concentration of ammonium nitrogen was 20 mg/L and the water was almost free of iron.

HRTs of nitrification and denitrification reactors were kept at 3 hours.

The initial sludge concentrations in both reactors were set at 2.5 mg/L.

Aeration rate to the nitrification reactor was maintained at 0.8 L/min.

The mixed rate in the denitrification reactors was 100 rpm.

The amounts of supplemental organic carbon in form of methanol, ethanol and glucose were added to the denitrification reactors in accordance with the ratio of C: N = 1: 1.

2.4 Analytical methods

Flow rate, pH, DO and alkalinity were monitored every 2 days and SS, NH₄-N, NO₃-N and NO₂-N levels were determined in the effluent. The pH and DO were measured by using a Mettler Toledo-320 pH meter (Switzerland) and UC-12 Digital DO/ O₂/ Temp. Meter (TOA, Ltd., Japan), respectively. Analyses of NH₄-N, NO₃-N, NO₂-N, MLSS, SS and alkalinity were performed according to Standard Methods for the examination of Water and Wastewater.

3. Results and discussion

3.1 Batch assay

Fig. 3.1 shows the time courses of $\text{NO}_3\text{-N}$ concentrations during the batch assay. Methanol, ethanol and glucose were used as a supplemental carbon sources to active the denitrification reaction. As shown, the denitrification rate was processed rapidly with glucose and ethanol during first 3 hours. It seems that ethanol and glucose are effective carbon sources for the denitrification process in comparison with methanol.

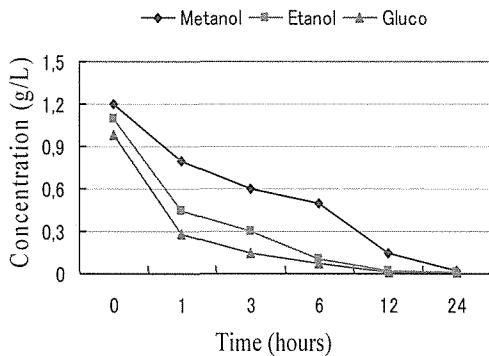


Fig. 3.1 Time course of $\text{NO}_3\text{-N}$ concentration during the batch tests.

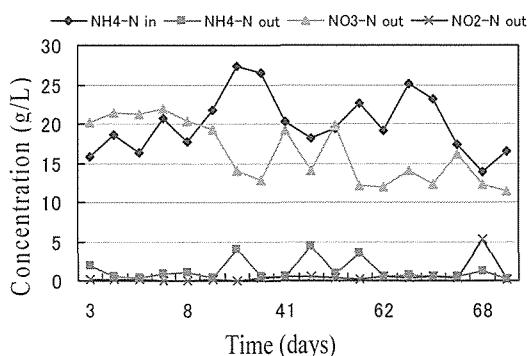


Fig. 3.2 Nitrogen concentration in effluent of nitrification reactor.

3.2 Continuous flow assay

3.2.1 Nitrification reactor

Fig. 3.2 shows the time courses of nitrogen concentrations in the effluent of nitrification reactor. The results revealed that, the reactor was able to nitrify about 20 mg/L of ammonium in the influent to the nitrate of 18 mg/L in average.

The alkalinity of raw groundwater in Phap Van areas was high of about 250 mg/L (as CaCO_3) and it was decreased to about 150 mg/l in the effluent (**Fig. 3.3**). This excessive amount of alkalinity could ensure the neutral pH in the reactor. This condition is good for growth of nitrifying bacteria.

The nitrification efficiency of the reactor is show in **Fig.3.4**. These results of continuous-flow experiments consistently demonstrated a high rate of nitrification using the BL material. The reactor was achieved 95~99% nitrification efficiencies with 20 mg/l of ammonium concentration in the influent.

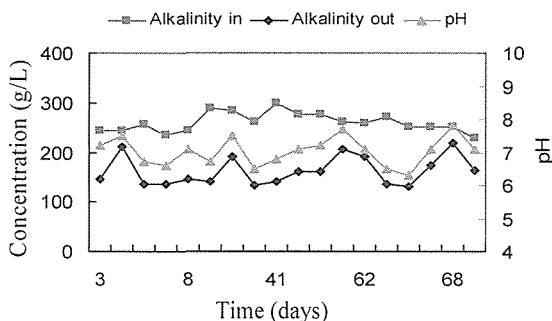


Fig. 3.3 Alkalinity and pH in nitrification reactor

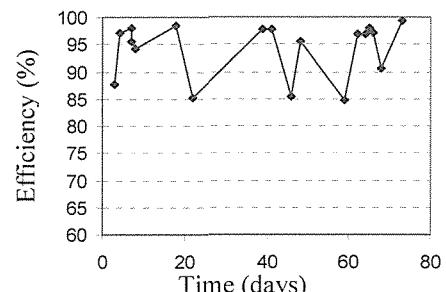


Fig 3.4 Nitrification efficiency.

3.2.1 Denitrification reactor

The concentrations of $\text{NO}_3\text{-N}$ in the influent and effluents of denitrification reactors, which used methanol, ethanol and glucose as a supplemental carbon sources, are shown in Fig. 3.5. Low $\text{NO}_3\text{-N}$ concentrations of about 5 and 2.5 mg/L were obtained in the effluents from the reactors which used ethanol and glucose, respectively. The average values of nitrate removal efficiencies of the reactors with methanol, ethanol and glucose were 58, 70 and 80%, respectively Fig. 3.6.

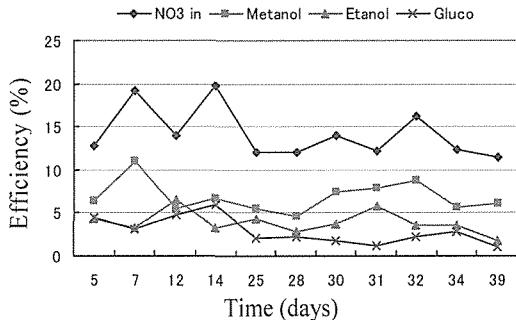


Fig 3.5 $\text{NO}_3\text{-N}$ in effluent of denitrification reactor

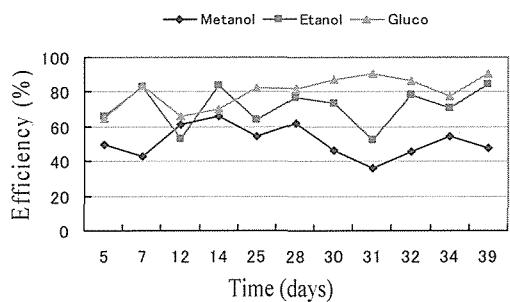


Fig. 3.6 Nitrate removal efficiencies with different carbon sources.

Fig. 3.7 shows the time course of $\text{NH}_4\text{-N}$ in the system's effluents. Results revealed that, the concentration of ammonium in the effluent from the reactor using methanol was about 1.5 mg/L, which can meet the Vietnamese standard. And it was about 0.8 mg/L for glucose and ethanol.

The comparative results of the continuous-flow nitrification-denitrification systems using different carbon sources demonstrated that use of ethanol and glucose as a supplemental carbon sources could achieve a good nitrogen removal efficiency of 93% and 97%, respectively, and it was higher in comparison with methanol (Fig. 3.8).

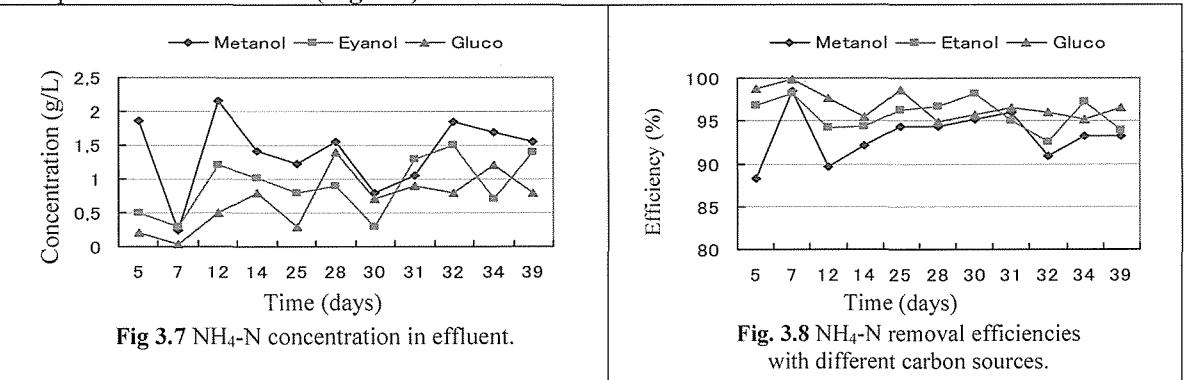


Fig 3.7 $\text{NH}_4\text{-N}$ concentration in effluent.

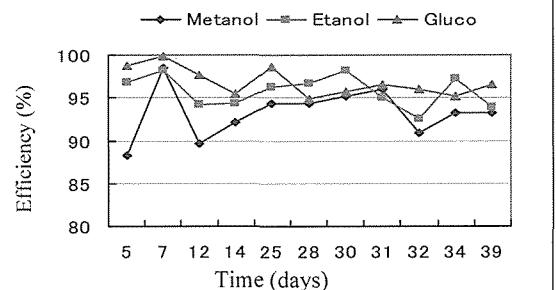


Fig. 3.8 $\text{NH}_4\text{-N}$ removal efficiencies with different carbon sources.

4. Conclusion

The following conclusions have been drawn from the experimental results obtained during the batch assay with synthetic groundwater and the continuous flow experiment with ammonium contaminated raw groundwater at Phap Van water treatment plant.

❖ The use of biological nitrification-denitrification system using a BL material as a biomass attachment medium has demonstrated potential for removal of ammonium from polluted groundwater in Hanoi. The system can play an effective role in providing safe drinking water.

- ❖ Ethanol and glucose are effective for the denitrification process and ethanol could be a good material due its economical mean in comparison with glucose. Therefore, the use of ethanol can be recommended as a supplemental carbon source for the treatment of ammonium contaminated groundwater in southern part of Hanoi.

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