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Comparison of organic and inorganic packing materials in the removal of ammonia gas in biofilters

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Abstract

Two organic and two inorganic packing materials were compared with regard to the removal of ammonia gas in a biofilter inoculated with night-soil sludge. By gradually increasing the inlet load of ammonia, the complete removal capacity, which was defined as the inlet load of ammonia that was completely removed, and the maximum removal capacity of ammonia, which was the value when the removal capacity leveled off for each packing material, were estimated. Both values which were based on a unit volume of packing material, were higher for organic packing materials than inorganic ones. By using kinetic analysis, the maximum removal rate of ammonia, V_m, and the saturation constant, K_s, were determined for all packing materials and the values of V_m for organic packing materials were found to be larger. By using the kinetic parameters, the removal rates for ammonia were compared among the four packing materials, and the organic packing materials showed superior performance for the removal of ammonia in the concentration range of 0–300 ppm as compared to inorganic packing materials. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Organic packing material; Inorganic packing material; Ammonia; Biofilter

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1. Introduction

Ammonia gas is a notable malodorous gas among volatile compounds. For the treatment of malodorous gases, physical and/or chemical methods have been popularly

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used. Although the efficiency of these methods is generally satisfactory, their maintenance and operation costs are high, and thus biological methods have attracted attention as alternative methods. Among the biological methods, a packed bed reactor system such as a biofilter has been reported to be an efficient method [1–7]. The selection of packing materials is an important factor in maintaining a high removal efficiency and a number of organic packing materials have been used successfully in biofilters [8–11]. When ammonia was supplied to peat, the ammonia was removed mainly by adsorption on the packing material or absorption into water [8] and the biological removal rate for ammonia in biofilters was low. The concentration of ammonia exceeding 35 ppm was toxic to most of the microorganisms in peat biofilters [6]. The use of common heterotrophic bacteria to remove ammonia is inefficient because the demand for a nitrogen source in these bacteria is one-tenth of the demand for a carbon source, indicating that a continuous supply of carbon source is essential. Therefore, the utilization of nitrifying bacteria has the potential to realize high removability of ammonia because they require carbon dioxide as a carbon source instead of organic carbon. However, the critical load of ammonia on nitrifying bacteria has not been clearly determined.

On the other hand, inorganic packing materials which are inert to biological degradation have been proposed as stable packing materials in deodorization reactors [12–15]. However, simultaneous comparison of the performance of organic and inorganic packing materials under specific operation conditions has not been carried out.

In this paper, we compared four packing materials (two organic and two inorganic) with regard to the removal capacity of ammonia and kinetic analysis was applied to evaluate the removal rate of the gas for the packing materials.

2. Materials and methods

2.1. Packing material and sludge

The main properties of the two organic packing materials and two inorganic packing materials are shown in Table 1. As organic packing materials, fibrous peat (Takahashi Peat Moss, Hokkaido, Japan) and rock wool were used. The rock wool packing material

Table 1
Physicochemical characteristics of packing materials used in this study

Characteristics	Packing materials			
	Peat	Rock wool	Fuyolite	Ceramics
pH(−)	4.7	8.6	7.9	7.4
Packed density (g dry/l)	223	291	113	237
Main chemical composition (%)	Organic C (48.0) Ash (4.2)	Organic C (6.19) Ash (85.4)	SiO_4 (78.2) Ash (96.7)	SiO_4 (85.9) Ash (96.2)
Water loss rate[a] (−%/day)	4.5–12.7	2.4–8.3	14.9–37.4	9.4–25.1
Pressure drop[a] (mm H_2O/m)	26.4–459	38.5–468	26.7–386	25.8–382

[a] Minimum and maximum values in the range of space velocity from 100 h^{-1} to 400 h^{-1}.

is a commercial product (Nichiyasu, Tokyo, Japan) which consists of a mixture of rock wool and rice husks. The main constituent of the rock wool packing material was inorganic ash but 6% of organic carbon was detected. As discussed below, the rock wool packing material showed a similar nitrification capacity as peat, which is different from the case in conventional inorganic packing materials. Thus, we classified the rock wool packing material as an organic packing material. As inorganic packing materials, Fuyolite (Fuyo Perlite, Tokyo, Japan) and ceramics (Kubota, Tokyo, Japan) were used. Fuyolite is obsidian sintered to form a foam aggregate. Its average particle size is 10 mm, and bulk density is 0.12 g cm^{-3}. The ceramics are mainly silicate with an average particle size of 10 mm, and bulk density of 0.47 g cm^{-3}. Both have almost no organic carbon content, as shown in Table 1.

The pH of each packing material was measured after mixing 10 g of packing material with 90 g distilled water and centrifuging the mixture at $8000 \times g$ for 10 min. The packing materials, which were ground using a homogenizer (Excel-Auto, Nihon Seiki, Japan), were dried at 105°C for 24 h and subjected to elemental analysis. The rate of water loss was measured as follows: each packing material, dried beforehand, was packed into a column up to 20 cm in height, sufficient water was supplied and the weight was measured to determine the initial water content (see Table 2). Then, air was supplied at a space velocity of 100 h^{-1} and the weight change was measured every 30 min. The rate of water loss was determined from the linear decrease in the weight. Simultaneously, the change in pressure drop was measured. Then, sufficient water was supplied to each packing material and similar experiments were conducted at different space velocities. From the data of rate of water loss, the time interval of the water supply to maintain the water content of packing materials at about 70% was determined.

In order to enhance the degradation rate of ammonia, inoculation of microorganisms was conducted. For seeding of the microorganisms, the sludge from a night-soil-treatment plant (22,000 mg l^{-1} of mixed liquor suspended solid (MLSS)) was sprayed onto each packing material at the ratio of 500 ml kg^{-1} packing material and then left to stand for one day to remove the attached drain.

Table 2
Experimental conditions of biofilters for ammonia removal

Experimental conditions	Packing materials			
	Peat	Rock wool	Fuyolite	Ceramics
Initial pH (−)	7.1	8.2	8.0	8.6
Initial dry weight of material (g)	77.9	102	39.4	82.8
Initial water content (%)	75	71	60	52
Initial packed volume (l)	0.35	0.35	0.35	0.35
Initial packed height (cm)	18	18	18	18
Flow rate (l/min)[a]	0.4–1.6	0.4–1.8	0.4–1.1	0.4–1.3
Space velocity (h^{-1})[a]	68–272	68–306	68–187	68–221
Inlet concentration (ppm)[a]	42–290	42–290	42–290	42–290
Nitrogen load (g N/kg dry material/day)[a]	0.19–4.52	0.19–3.89	0.19–6.17	0.19–3.91

[a] Maximum and minimum values.

2.2. Experimental apparatus

The laboratory-scale biofilter used is shown in Fig. 1. The four packing materials seeded with night soil sludge were packed in glass columns (5 cm in inner diameter, 50 cm in height) to a packing height of 18 cm. The ammonia gas from a cylinder was diluted with ambient compressed air and was supplied downwards to the columns. The inlet ammonia concentration was changed by controlling the air-flow rate. Thus, the load of ammonia into the biofilters was changed by controlling the inlet concentration and/or space velocity (= flow rate/packing volume).

2.3. Operating conditions

The initial experimental conditions are listed in Table 2. The experiment was carried out at a room temperature of 20–25°C. The concentration of ammonia in the inlet gas varied from 42–290 ppm, while the space velocity ranged from 68–272 h^{-1} for peat, 68–306 h^{-1} for rock wool and 51–187 h^{-1} for Fuyolite and ceramics. The initial pH of peat was adjusted to be neutral, using 5% Na_2CO_3. Water was splashed into the biofilter in the columns using peristaltic pumps (Fig. 1) once a day for organic packing materials and twice a day for inorganic packing materials to maintain the moisture content at about 70%. During this operation, the pH value of the drained water was measured. When the pH was alkaline, 30 ml of 0.5% HCl solution was added into the column. Then, the drained water was collected and its pH was measured. This procedure was repeated until the pH of the drain reached around 7. When the measured pH was more

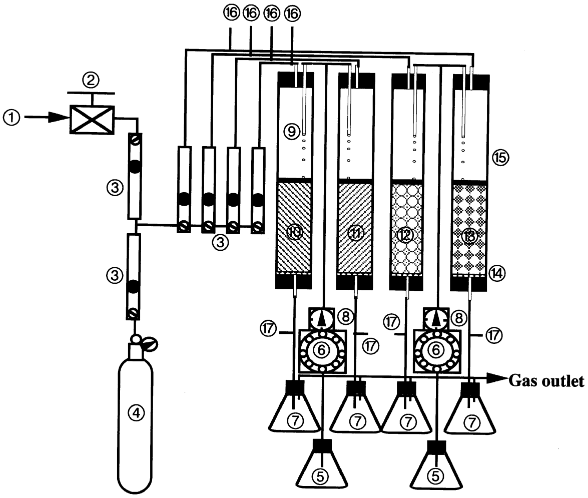

Fig. 1. Schematic diagram of a laboratory-scale biofilter for ammonia removal. (1) Pressurized air; (2) Regulator; (3) Flow meter; (4) Ammonia gas cylinder; (5) Water reservoir; (6) Peristaltic pump; (7) Drain trap; (8) Time controller; (9) Sprinkler; (10–13) Packing materials; (14) Saran net; (15) Column; (16) Inlet gas sampling port; (17) Outlet gas sampling port.

than 7.5% Na_2CO_3 solution was added and a similar procedure was carried out to control the pH to around 7.

2.4. Bacterial count

The cell number of nitrifying bacteria was estimated by the most probable number (MPN) method [16,17]. About 5 g wet weight of the packing material was sampled and homogenized in 95 ml of Alexander (AL) medium at 10,000 rpm for 10 min (EX-3, Nihon Seiki, Tokyo, Japan). AL medium contained 2.5 g $(NH_4)_2SO_4$, 0.5 g KH_2PO_4, 50 mg $MgSO_4$ $7H_2O$, 4 mg $CaCl_2$ $2H_2O$, and 0.1 mg Fe-EDTA per liter, at pH 8.0–8.2 [18]. The homogenized suspension was diluted with AL medium, then 0.5 ml of the suspended solution with different dilution ratios was transferred to 4.5 ml AL medium in 18 cm test tubes, and incubated at 30°C at 120 spm in the dark for 3 weeks. At the end of the incubation period, each tube was scored by adding an indicator (2.2 g of diphenylamine in 100 ml of concentrated H_2SO_4) to test the presence of nitrite and/or nitrate. A blue color reaction indicated that nitrite and nitrate were formed, and the tube was scored positive. The absence of a blue color was scored negative. By referring to the MPN table, the positive number can be used to estimate the population cell number [16,17].

2.5. Analysis

The inlet and outlet ammonia concentrations in the biofilter were measured using ammonia gas detection tubes (Gastec, Tokyo, Japan). The lower detection limit of the tubes was 0.25 ppm and the error of measurement was $\pm 5\%$. To test the presence of ammonium ion, nitrate and nitrite in the drained water, a merckoquant test strip (Merck KGaA, Darmstadt, Germany) was used for each chemical.

3. Results and discussion

3.1. Physicochemical properties of each packing material

Physicochemical properties of each packing material are shown in Table 1. Elemental analysis showed that peat and rock wool as organic packing materials contained organic carbon, while Fuyolite and ceramics as inorganic packing materials were devoid of carbon. The pH of peat was acidic mainly due to the presence of humic substances, while the other packing materials were neutral or alkaline. Rates of water loss for peat and rock wool were -4.5 and -2.4% day^{-1} at the space velocity of 100 h^{-1}, and for Fuyolite and ceramics, they were -14.9 and -9.4% day^{-1}, respectively. When the space velocity was increased to 400 h^{-1}, the loss of water from inorganic packing materials was 2–3-fold that for organic packing materials. These values indicate that the loss of water from inorganic packing materials was significantly higher than that from organic packing materials. The pressure drop for organic packing materials was higher at a space velocity of 100 h^{-1} mainly due to the fine particles and pressurized packing

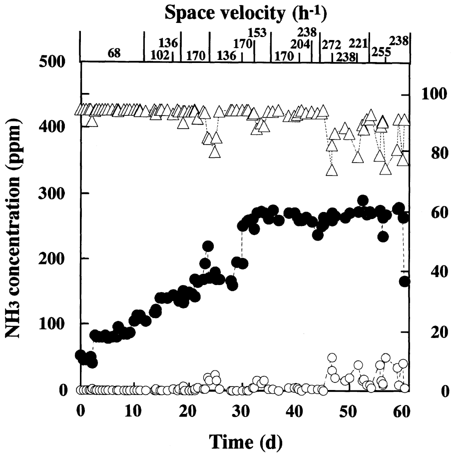

Fig. 2. Change in inlet concentration (●) and outlet concentration (○) of ammonia and removal ratio (△) in a biofilter with peat as a packing material.

during operation. At a higher space velocity, the increase in pressure drop for organic packing materials was larger.

3.2. Ammonia removal

The changes in the inlet and outlet ammonia concentrations, and removal ratio, at each space velocity during 60-day operation are shown in Fig. 2 for peat, Fig. 4 for rock wool, Fig. 6 for Fuyolite and Fig. 8 for ceramics. The changes in pH and nitrogenous compounds in the drained water are shown in Fig. 3 for peat, Fig. 5 for rock wool, Fig. 7 for Fuyolite and Fig. 9 for ceramics.

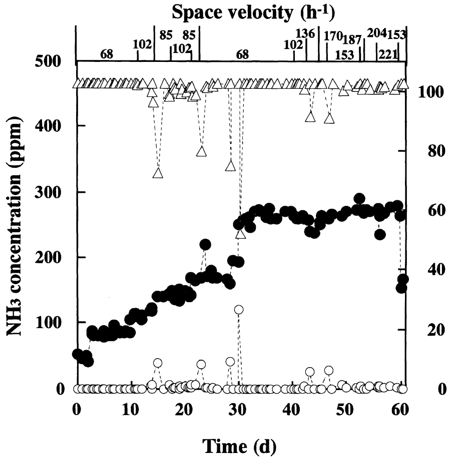

Fig. 3. Change in inlet concentration (●) and outlet concentration (○) of ammonia and removal ratio (△) in a biofilter with ceramics as a packing material.

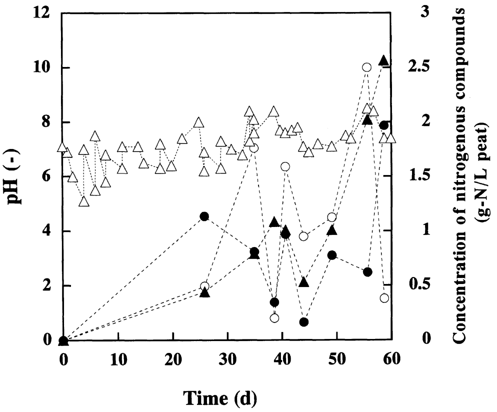

Fig. 4. Change in pH (△), and concentrations of NH_4^+ (●), NO_2^- (▲) and NO_3^- (○) in a drain of a biofilter with peat as a packing material.

In the initial 20 days of the operation, no ammonia at the outlet of peat was detected and the removal ratio was maintained at 100%. Then, when the space velocity of peat was raised from 136 to 170 h^{-1} and the inlet ammonia concentration was raised from 180 to 220 ppm, ammonia was detected at the outlet. Thus, the inlet ammonia concentration was decreased to 180 ppm, the space velocity was reduced to 136 h^{-1}, and 100% removal ratio was resumed (Fig. 2). Subsequently, inlet ammonia concentration was fixed at around 280 ppm and the space velocity was varied to assess the critical load of ammonia. The removal ratio for rock wool was similar to that of peat (Fig. 4). During the initial 20 days, no ammonia was detected at the outlet. On the 25th day, the decrease in the removal ratio was obvious, which was reflected as a pH rise to about 8

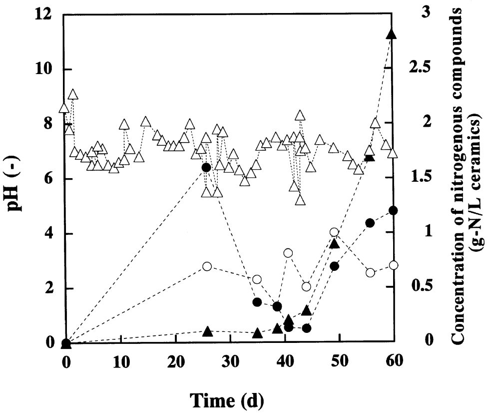

Fig. 5. Change in pH (△) and concentrations of NH_4^+ (●), NO_2^- (▲) and NO_3^- (○) in a drain of a biofilter with ceramics as a packing material.

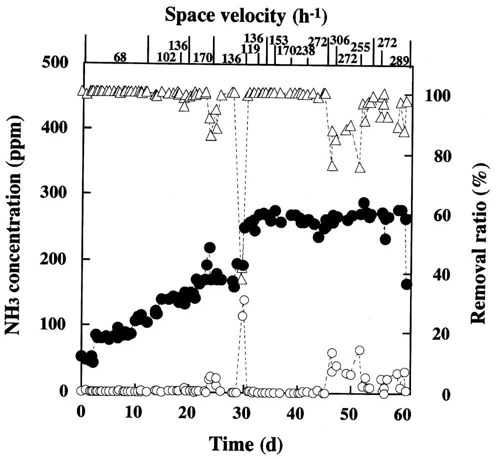

Fig. 6. Change in inlet concentration (●) and outlet concentration (○) of ammonia and removal ratio (△) in a biofilter with rock wool as a packing material.

(Fig. 5), and the space velocity was reduced. On the 30th day, the removal ratio declined to 40% mainly due to the quick increase in inlet ammonia concentration from 180 to 240 ppm. The 100% removal ratio was resumed by decreasing the space velocity and adjusting the pH. Subsequently, at the inlet ammonia concentration of about 280 ppm, the space velocity was varied.

Fuyolite showed a decrease in the removal ratio on the 15th day at space velocity of 102 h^{-1} and a 100% removal ratio at an inlet concentration of 280 ppm was established at considerably lower space velocity than in the case of peat or rock wool. When the space velocity was 170 h^{-1}, the removal ratio fluctuation was significant after 48 days of operation (Fig. 6). Similar fluctuations occurred at 250–300 h^{-1} for peat and rock

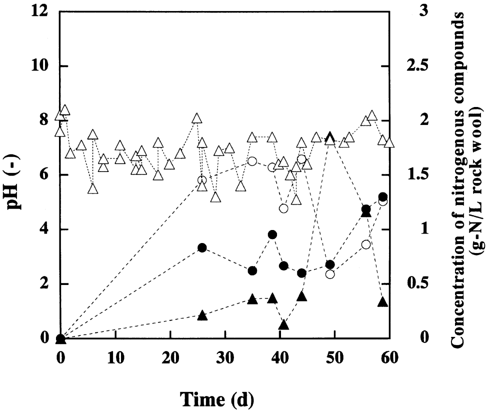

Fig. 7. Change in pH (△) and concentrations of NH_4^+ (●), NO_2^- (▲) and NO_3^- (○) in a drain of a biofilter with rock wool as a packing material.

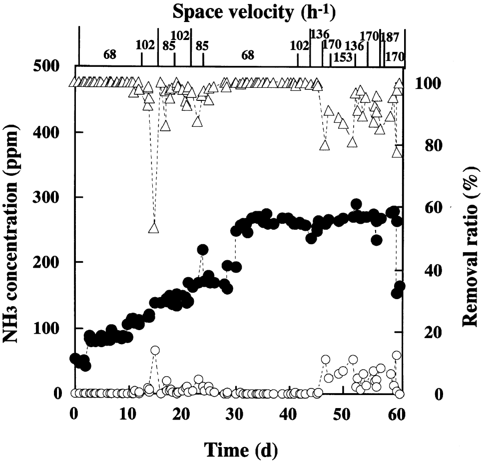

Fig. 8. Change in inlet concentration (●) and outlet concentration (○) of ammonia and removal ratio (△) in a biofilter with Fuyolite as a packing material.

wool (Figs. 2 and 4). Ceramics showed a similar behavior to Fuyolite until 40 days. After 40 days, the removal ratio was constant.

The removal capacity of ammonia for each packing material is shown by the change in the accumulation of nitrogenous compounds in each packing material (Figs. 3, 5, 7 and 9). The concentrations of the nitrogenous compounds were expressed as the concentration per unit volume of each packing material. The increase in nitrite (NO_2^-) and nitrate (NO_3^-) concentrations was significantly higher for organic packing materials than for inorganic packing materials. From those four figures, the values of net removal of NH_3–N as unit of g N/l packing material for 60 days were calculated 22.1, 23.5,

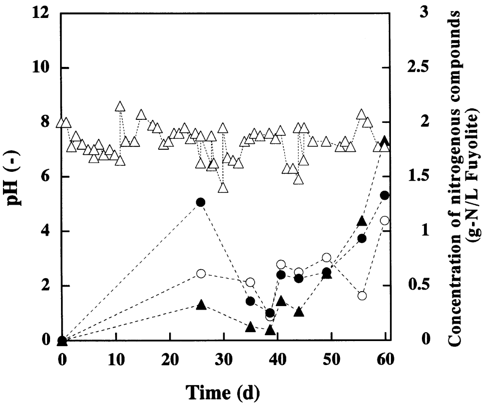

Fig. 9. Change in pH (△) and concentrations of NH_4^+ (●), NO_2^- (▲) and NO_3^- (○) in a drain of a biofilter with Fuyolite as a packing material.

10.5 and 13.9 for peat, rock wool, Fuyolite and ceramics, respectively. The sums of nitrite and nitrate accumulated on each packing material were 18.5, 15, 9.6 and 11.4 (g N/l packing material) for peat, rock wool, Fuyolite and ceramics, respectively. These data indicate that removed amount of NH_3–N and accumulated amount of nitrification products are both significantly higher in organic packing materials.

3.3. Microorganisms

The initial nitrifying bacteria number in the original sludge was determined as 4.1×10^4 cells ml^{-1} by the MPN method. After the sludge was sprayed onto each packing material, the concentration was of the order of 10^5 cells/g dry material. After 60 days of the experiment, nitrifying bacteria on the peat and the rock wool increased to about 1.1×10^8 cells/g dry material and to 3.3×10^7 cells/g dry material for Fuyolite and ceramics, respectively. The significant increase in the nitrifying bacteria on each packing material was obvious. Although the MPN method gives a statistical estimation of chemoautotrophic nitrifiers, the difference in the cell numbers shown above between organic and inorganic packing materials can be significant. This difference can be explained by the fact that a higher load can be imposed on organic packing materials and a higher accumulation of NO_3^- was observed. This suggests that organic packing materials provide a more suitable environment for nitrifying bacteria than inorganic packing materials, although the details of the interaction between packing materials and nitrifying bacteria are not clear.

3.4. Removability of ammonia

The relationship between the load to each packing material and the removal capacity is shown in Fig. 10. The complete removal capacity was defined as the inlet load of

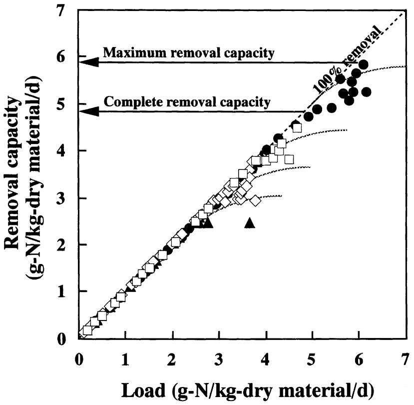

Fig. 10. Relationship between removal capacity and load of ammonia for biofilters with four packing materials. □: peat, ◇: rock wool, ●: Fuyolite, ▲: ceramics. ←: maximum removal capacity, ←: complete removal capacity.

Table 3
Maximum and complete removal capacities of ammonia by biofilters using four packing materials

Packing materials	Maximum removal capacity		Complete removal capacity	
	g N/kg dry material/day	g N/m^3/day	g N/kg dry material/day	g N/m^3/day
Peat	4.5	1.0×10^3	3.2	7.2×10^2
Rock wool	4.0	1.2×10^3	2.8	8.1×10^2
Fuyolite	6.0	6.8×10^2	4.7	5.3×10^2
Ceramics	3.9	9.2×10^2	2.4	5.7×10^2

ammonia that was completely removed and thus no outlet concentration is detected, while the maximum removal capacity was defined as the value when the removal capacity leveled off. The values for the complete removal capacity and maximum removal capacity are summarized in Table 3. On the basis of weight, the maximum removal capacity and complete removal capacity were the highest for Fuyolite. However, on the basis of volume, both values for organic packing materials are larger than those for inorganic packing materials. This difference is mainly due to the difference in the packing density of each packing material as the packed volume and packed height of each column were equal at the start of experiment. From the engineering viewpoint, the compactness of the reactor is of primary concern. Thus, considering a compact reactor design, the use of organic packing materials is preferable. The data of the maximum removal capacity of each packing material can be applied in the design of a reactor. The complete removal load of ammonia was previously reported to be 0.16–0.17 g N/kg dry peat/day [8,10] when nitrifying sludge was inoculated onto peat. In this experiment, the value was about 20 times larger for all packing materials. This may be primarily due to the careful control of the inlet concentration of ammonia, space velocity, and pH. In a previous experiment [10] in which the operation period was shorter, the overload of ammonia was calculated from the reported data as 16×10^2 g N/m^3/day or 1.8 g N/kg/day and the inlet concentrations were around 50 ppm. The maximum removal capacities listed in Table 3 were significantly larger than the reported values. Therefore, the values listed in Table 3 indicate the maximum capacity of nitrification when a natural microbial community of nitrifying microorganisms was applied to biofilters.

3.5. Kinetic analysis

After 60 days of operation, the data for the kinetic analysis of each packing material were obtained from the transient increase in the ammonia load which was conducted by increasing the concentration of the ammonia from 302 to 408 ppm in the range of space velocity of 204 and 450 h^{-1} for peat, from 302 to 353 ppm in the range of 272 and 408 h^{-1} for rock wool, from 180 to 360 ppm in the range of 153 and 221 h^{-1} for Fuyolite, and from 216 to 360 ppm in the range of 187 and 216 h^{-1} for ceramics. The removal rate of ammonia in a biofilter was assessed in similar ways as previously reported [8,12,15]. The plug flow of ammonia gas was assumed in the biofilter column. It was also assumed that no mass transfer or diffusion of ammonia gas was limiting because

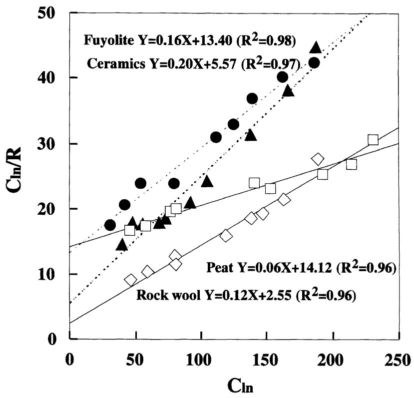

Fig. 11. Kinetic analysis of ammonia removal by a biofilter with four packing materials. The correlation equation for each packing material is shown together with correlation coefficient, R^2. □: peat, ◇: rock wool, ●: Fuyolite, ▲: ceramics.

high inlet ammonia gas was supplied during the experiment. Thus, the Michaelis–Menten-type equation was applied as

$$\frac{-\mathrm{d}C}{\mathrm{d}l} = \frac{V_\mathrm{m}C}{K_\mathrm{s}+C}\left(\frac{S_\mathrm{a}}{F}\right)\alpha, \tag{1}$$

$$\frac{-\mathrm{d}C}{\mathrm{d}l} = \frac{V_\mathrm{m}C}{K_\mathrm{s}+C}\left(\frac{1}{L\cdot \mathrm{SV}}\right)\alpha, \tag{2}$$

where C: ammonia concentration (ppm); l: length of column (m); V_m: maximum removal rate (g N/kg dry material/day); K_s: saturation constant (ppm); S_a: cross-section of column (m^2); F: gas flow rate (m^3/ day); L: height of packed peat (m); SV: space velocity (day^{-1}) $= F\,S_\mathrm{a}^{-1}L^{-1}$; α: conversion coefficient (kg dry material/g N).

The conversion coefficient, a, defined by Eq. (3), was used to convert the units of concentration to ppm.

$$\alpha = \frac{\left[22.4 + \dfrac{273+T}{273}\right]\times 10^6}{14\times 1000}\,\frac{W}{V}, \tag{3}$$

where T: temperature (°C); W: dry weight of packing material (kg); V: volume of packing material (m^3); 14: the atomic weight of nitrogen.

Integrating Eq. (2) under the condition of $C = C_0$ at $l = 0$ and $C = C_\mathrm{e}$ at $l = L$, we obtain

$$\frac{\alpha}{\mathrm{SV}(C_0 - C_\mathrm{e})} = \frac{K_\mathrm{s}}{V_\mathrm{m}}\,\frac{1}{\dfrac{(C_0 - C_\mathrm{e})}{\ln(C_0/C_\mathrm{e})}} + \frac{1}{V_\mathrm{m}}. \tag{4}$$

Table 4
Maximum removal rate (V_m) and saturation constant (K_s) of ammonia estimated from kinetic analysis in the biofilters using four packing materials

Packing materials	Maximum removal rate, V_m		Saturation constant, K_s (ppm)
	g N/kg dry material/day	g N/m^3/day	
Peat	16.7	3.7×10^3	235
Rock wool	8.3	2.4×10^3	21
Fuyolite	6.3	7.1×10^2	84
Ceramics	5.0	1.2×10^2	28

Setting $R = SV(C_0 - C_e)/\alpha$ and $C_{ln} = (C_o - C_e)/\ln(C_o/C_e)$, Eq. (4) is simplified to:

$$\frac{C_{ln}}{R} = \frac{K_s}{V_m} + \frac{C_{ln}}{V_m}. \tag{5}$$

The relation between C_{ln}/R and C_{ln} is shown in Fig. 11 and the correlation equations are included in the figure together with the correlation coefficients. The maximum removal rate, V_m, and the saturated constant, K_s, are listed in Table 4. The values of V_m of ammonia for organic packing materials are larger than those of inorganic packing materials.

As the overall reaction rate for ammonia removal is determined by both V_m and K_s, which are dependent on the packing materials, the kinetic equation using V_m and K_s in Eq. (1) for each packing material was compared, as shown in Fig. 12. The reaction rate of rock wool is superior to that of any other packing material in the range of ammonia concentration between 0 to 190 ppm. Beyond 190 ppm, peat has a superior performance to any of the other packing materials. The difference between Fuyolite and ceramics was small. When the concentration was below 60 ppm, ceramics have a superior performance to peat and Fuyolite. The advantage of organic packing materials is that organic

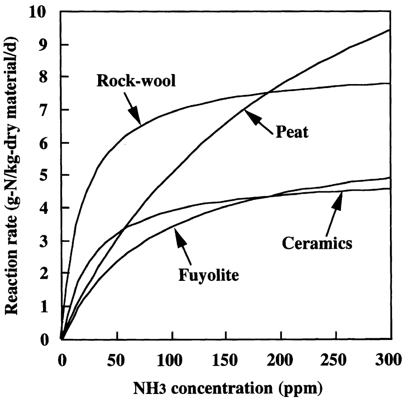

Fig. 12. Relationship between reaction rate of ammonia and ammonia concentration in biofilters with four packing materials of peat, rock wool, Fuyolite and ceramics.

packing materials act as inorganic and/or organic nutrients for microorganisms and have a buffering capacity to pH change. However, inorganic packing materials have a weak buffering capacity against pH fluctuation and are devoid of nutritional compounds that support the metabolism of microorganisms. In this experiment, the frequency of supply of water to adjust pH for inorganic packing materials was about 2–3 times more than that for organic packing materials. Thus, it can be concluded that organic packing materials are suitable for the removal of ammonia because the change in pH due to the metabolized products by organic packing materials can be alleviated and pH control is easier. However, in the case of long-term operation of more than few years, special measures must be taken to prevent the increase in pressure drop for organic packing materials which was primarily caused by their decomposition and change in particle size.

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