

# Granulated Coal Ash-Used Filter Design for Efficient Removal of Nutrient Salts

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**Abstract-** To date, granulated coal ash (GCA, a by-product from coal fired-power plants) has been widely used for improving sediment and water quality, such as removals of nutrient salts and hydrogen sulfide. These removal capacities of GCA lead us to consider that GCA can be used as a filter material in wastewater treatments. This study is aiming at examining the potential of using GCA as a filter material in wastewater treatments. One of our objectives is to examine how to use GCA for efficient removal of nutrient salts. Another objective is to propose filter design and operation of using GCA for removing nutrient salts effectively. Laboratory experiments were conducted under different conditions, such as different concentrations of nutrient salts and retention times. The experimental results showed a higher removal rate of ammonium ion when polluted water was retained in GCA filter compared to the circulating polluted water through the GCA filter. This was observed when the retention time shorter than 180 min. However, the removal rate was similar when the retention time was longer than 180 min. A higher solution pH in the retention condition was considered to partly contribute to the higher removal rate. Moreover, the absorbed quantity increased with an increase in the concentrations of nutrient salts. This pointed out that the removal of nutrient salts by GCA depends on the concentrations of nutrient salts present in the solution. According to these results, design of GCA filter and its operation were proposed. The GCA filter was divided into two different parts. The polluted water was retained in the first part for 90 min, then discharging to the second part for 90 min-retention. Compared with the filter without division (180 min-retention), 1.33-fold increases in  $\text{NH}_4^+$  and 1.55-fold increases in  $\text{PO}_4^{3-}$  removals were obtained owing to our proposed filter design and operation.

**Keywords-** Granulated Coal Ash; Filter Design; Removal; Nutrient Salts; Retention

## I. INTRODUCTION

The annual production of fly ash is approximately 500 million tones which is at 75-80% of the total ash produced, while the current utilization rate of ash is still low (the world average only amounts to 16% of the total ash) [1]. Fly ash has been used in many different fields, such as agricultural soil modification, roadway and pavement construction, cement and concrete products, ceramic materials, soil and water treatment, and catalytic applications [2-8]. Details can be found in the report by Belviso [9]. The utilization rate of fly ash has increased; however, it is up to 25% in recent years [2]. Therefore, the increasing utilization rate of fly ash is required.

For the purpose of increasing the utilization rate of fly ash, our research group has developed granulated coal ash (GCA) for improving sediment and water quality in littoral regions. GCA refers to Hi-beads that are a commercial product of Energia Eco Material Co. Inc. (Japan). GCA is made by granulating fly ash from coal fired-power plants with a small amount of cement and water as binders. According to Asaoka et al. [10], the primary components of GCA are  $\text{SiO}_2$  (395 g/kg),  $\text{Al}_2\text{O}_3$  (126 g/kg),  $\text{CaO}$  (55.4 g/kg), and  $\text{Fe}_2\text{O}_3$  (22.5 g/kg). The specific surface area of GCA is 21.1  $\text{m}^2/\text{g}$ .

GCA has been recommended as an absorbent. Asaoka et al. [11] mixed sediment with GCA and examined the elution of nutrient salts and  $\text{H}_2\text{S}$  from the sediment into overlying water. They reported that the mixing GCA with the sediment could reduce phosphate ( $\text{PO}_4^{3-}$ ) releasing flux from the sediment into overlying water by 37-44% compared to the case without using GCA. In the study by Touch et al. [12], GCA was mixed with sediment and changes in the quality of the sediment pore water and overlying water were examined. The authors reported decreases in  $\text{H}_2\text{S}$ , Manganese (Mn), ammonium ( $\text{NH}_4^+$ ) and  $\text{PO}_4^{3-}$  in the sediment pore water were 99%, 96%, 60% and 93%, respectively; and those in the overlying water were 99%, 96%, 65% and 67%, respectively. Thus, GCA has been widely used for improving sediment and water quality in littoral regions. Kim et al. [13] aimed to improve the bottom water quality of Kaita Bay (the innermost area of Hiroshima Bay, Japan) by scattering GCA on the seafloor. Their results suggested that the scattered GCA layer could reduce the release of nutrient salts and  $\text{H}_2\text{S}$  from the sea bottom sediment to bottom water. Yamamoto et al. [14] also conducted a pilot study for improving the marine water quality at oyster farming sites using GCA. The authors also demonstrated that scattered GCA layer could effectively reduce the release of nutrient salts and  $\text{H}_2\text{S}$  from the sea bottom sediment to bottom water. They also reported that the removal of such substances improved the benthic habitat environment by increasing the abundance and species richness of benthic microalgae organisms present in the surface sediment.

If GCA can be used to solve different environmental issues, the using amount of GCA will increase. Recently, wastewater discharge associated with combined sewer over flows increases in proportion to the population growth and urban development. However, wastewater treatment plants are unable to treat all amounts of the increased wastewater discharges.

This study aims to examine the potential of using GCA as a filter material in wastewater treatments. Laboratory experiments are conducted under different conditions, such as different concentrations of nutrient salts and retention times. These allow us to understand how to use GCA for efficient removal of nutrient salts, and to propose a filter design of using GCA and its operation to effectively remove nutrient salts. It is expected that our achievements may contribute to efficient wastewater treatments in combined sewer overflows. This in turn can lead to the increasing utilization rate of fly ash (the main component of GCA) produced from coal-fired power plants.

## II. MATERIALS AND METHODS

### A. Materials Used in the Experiments

The GCA used in the experiments was obtained from Energia Eco Material Co. Inc. GCA particles range from coarse sand to gravel (Fig. 1a). The diameter of GCA is up to 40 mm and the mean diameter is approximately 20 mm. Moreover, GCA is a porous material which has a small specific gravity. The particle density of GCA ranges from 0.8 to 1.1 t/m<sup>3</sup> in a dry condition, and 1.0 to 1.4 t/m<sup>3</sup> in a wet condition [15]. Asaoka et al. [10] have reported that the environmental regulated substances dissolved from GCA are obviously below the standard levels of polluted soils in Japan. Saito et al. [16] have reported that a particle of GCA comprised micropores with a diameter range of 2-100 nm, and has a porosity of 40%. GCA was sieved before being used in the experiments. As a result, the GCA with a diameter range of 4.25-26.5 mm was used as the filter material in this study. The GCA was first washed with tap water and then it was air dried prior to use in the experiments.



Fig. 1 Physical appearance of GCA (a) and fly ash (b).

Different polluted waters (PW1, PW2, and PW3) used in the experiments were made by the following procedures: (i) the sea bottom sediment collected from a water body exposed to wastewater discharge was mixed with tap water at a volume rate of 1 (sediment):2 (tap water) in a 30 L-clean plastic bucket; (ii) the bucket was placed in an incubator (Fukushima, FMU-404I) at 30 °C for 2 weeks (the sample in the bucket was mixed every 3 days); (iii) the bucket was mixed at a day before the experiment and settled for 24 h. The overlying water, i.e., polluted water (PW1, PW2, or PW3), was then extracted from the bucket and used in the experiments. On the other hand, PW4 was made by mixing KH<sub>2</sub>PO<sub>4</sub> and NH<sub>4</sub>Cl with deionized water. The mixing rate was decided based on the concentration that will be used in the experiments. The initial quality of each polluted water is listed in Table 1. In the table, the pH and ORP were measured by inserting pH/ORP meters (Horiba, D-50) into the polluted water directly. The EC and DO concentrations were measured by inserting an EC meter (Horiba, D-74) and a DO meter (WTW, FDO925) into the polluted water, respectively. The NH<sub>4</sub><sup>+</sup>, PO<sub>4</sub><sup>3-</sup> and SO<sub>4</sub><sup>2-</sup> concentrations were measured using a digital water quality analyzer (Kyoritsu Chemical-Check Lab. Corp., A9000). The Ca<sup>2+</sup> and Na<sup>+</sup> concentrations were measured using Horiba-laqua twin electrodes. For measuring dissolved ions, the polluted water was filtrated through 45µm-membrance before it was used. Double measurements of each parameter were conducted and the average value was used in discussion.

TABLE 1 THE INITIAL QUALITY OF THE POLLUTED WATER USED IN THE EXPERIMENTS

Parameter	Unit	Polluted water (PW)				Measurement error (n=2)
		PW1	PW2	PW3	PW4	
pH		7.64	7.65	7.54	6.47	±0.03
Redox potential (ORP)	mV vs. Ag/AgCl saturated KCl	-370	-372	-361	285	±3
Electrical conductivity (EC)	µS/cm	15410	10140	19110	190	±10
Dissolved oxygen (DO) concentration	mg/L	0.05	0.01	0.01	N/A	±0.1
Ammonium (NH <sub>4</sub> <sup>+</sup> ) concentration	mg/L	52.00	31.25	41.50	25.25	±0.02
Phosphate (PO <sub>4</sub> <sup>3-</sup> ) concentration	mg/L	17.75	12.25	26.25	27.00	±0.04
Calcium (Ca <sup>2+</sup> ) concentration	mg/L	86.50	66.50	160	3.5	±1
Sodium (Na <sup>+</sup> ) concentration	mg/L	3000	1900	3850	3.5	±20
Sulfate (SO <sub>4</sub> <sup>2-</sup> ) concentration	mg/L	16	10	10	10	±4

### B. The Experimental Procedures and Conditions

The experiments were conducted with three different filter designs (FD1, FD2, and FD3), as shown in Fig. 2. Each filter was made from a cylindrical PVC with an inner diameter of 100 mm. The filter length was 200 mm for either FD1 or FD2, and

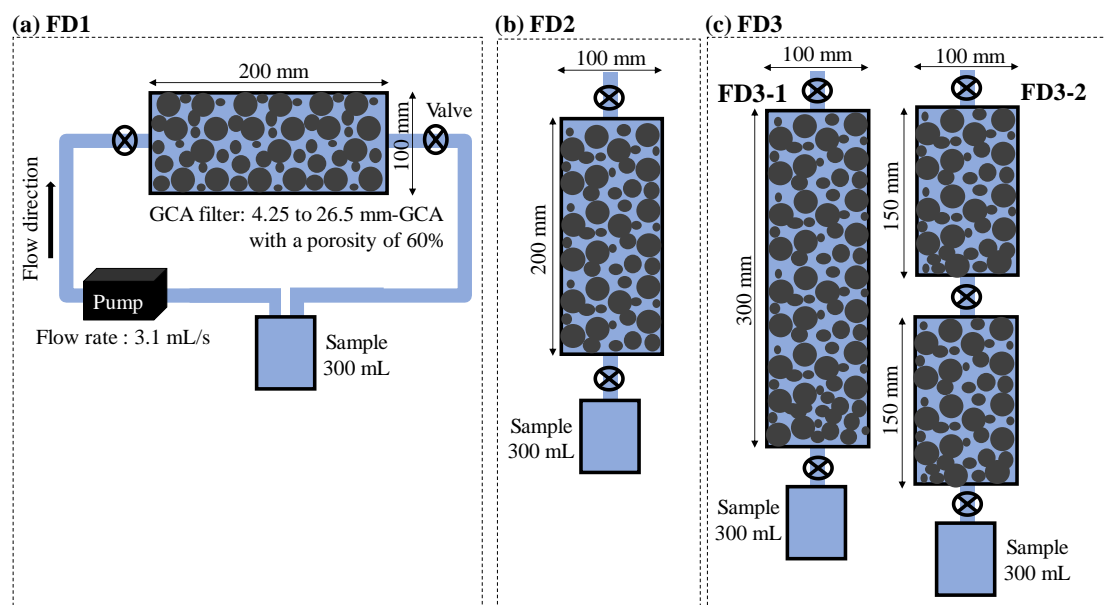


Fig. 2 Designs of the GCA filters used in the experiments.

150 mm or 300 mm for FD3. The cap with a valve was used to close the filter. The filter was filled with the washed and dried GCA with a porosity of 60% (without compaction) on average, and the polluted water was then poured into the filter.

The experiments were conducted with the conditions listed in Table 2. In FD1 (Cases 1-6), PW1 was circulated in the GCA filter at a flow rate of 3.10 mL/s with different retention times (Table 2). The maximum retention time was decided based on our previous results (most of nutrient salts were removed within this retention time). The retention time was calculated based on Eq. 1. In other words, the filtration time was calculated to obtain the same retention time as that of FD2.

$$T = L \times A \times t / V \quad (1)$$

where,  $T$ : the retention time,  $L$ : the filter length,  $A$ : the filter-cross section,  $t$ : the filtration time, and  $V$ : the volume of polluted water used in the experiment.

In FD2 (Cases 7-12), PW2 or PW4 was poured and retained in the GCA filter at different retention times (Table 2). At the end of each retention time, the sample was analyzed following the procedures explained earlier, and the results were compared to those of the control (without GCA used) polluted water. To conduct a new experiment from one to another retention time, the GCA was replaced at the end of each retention time. In FD3, two cases of experiments were performed, i.e., FD3-1 and FD3-2. In FD3-1, the PW3 was retained in the GCA filter for 180 min, and then 300 mL of sample was extracted and analyzed. In FD3-2, the FD3-1 was divided into two equal parts (Fig. 2). Firstly, the PW3 was placed in the first part with a retention time of 90 min, and then discharging to the second part. The discharged water was retained in the second part for 90 min. Finally, 300 mL of the sample was extracted, and its quality was analyzed.

TABLE 2 CONDITIONS OF EXPERIMENTS FOR EXPLORING NUTRIENT SALTS REMOVAL BY GCA.

Conditions	Filter design	Polluted water	Flow rate (mL/s)	Retention time (min)	NH <sub>4</sub> <sup>+</sup> in polluted water without GCA (mg/L)	PO <sub>4</sub> <sup>3-</sup> in polluted water without GCA (mg/L)
Case 1	FD1 (circulation)	PW1	3.10±0.08	5	50.75	16.00
Case 2				15	55.00	14.50
Case 3				30	51.25	12.50
Case 4				60	58.25	14.50
Case 5				180	57.00	15.50
Case 6				360	60.75	17.25
Case 7	FD2 (retention)	PW2	/	15	33.00	12.25
Case 8				30	33.75	12.50
Case 9				60	33.75	12.25
Case 10				180	33.00	11.50
Case 11				360	32.75	12.75
Case 12		PW4		360	25.25	27.00
Case 13	FD3-1	PW3		180	21.5	10.00
Case 14	FD3-2			90 for each	45.00	26.00

## III. RESULTS AND DISCUSSION

## A. Removal Rate of Nutrient Salts with FD1 (Circulation) in Comparison with FD2 (Retention)

Filter design is very important in wastewater treatments. Either filtration (circulation) or retention is used in wastewater treatments. When using GCA, released ions from GCA also play an important role in the removal of nutrient salts. It is thought that the dissolution characteristics of GCA are different between circulation and retention conditions. Thus, the removal rates of nutrient salts due to the circulation or retention of polluted water in GCA filters are discussed in this study. Figure 3 shows the removal rates of  $\text{NH}_4^+$  and  $\text{PO}_4^{3-}$  calculated from the control (without GCA used) polluted water in FD1 in comparison with those in FD2. In our results, it was also confirmed that most of nutrient salts were removed within the retention time of 360 min (76% for  $\text{NH}_4^+$  and 91% for  $\text{PO}_4^{3-}$ ).

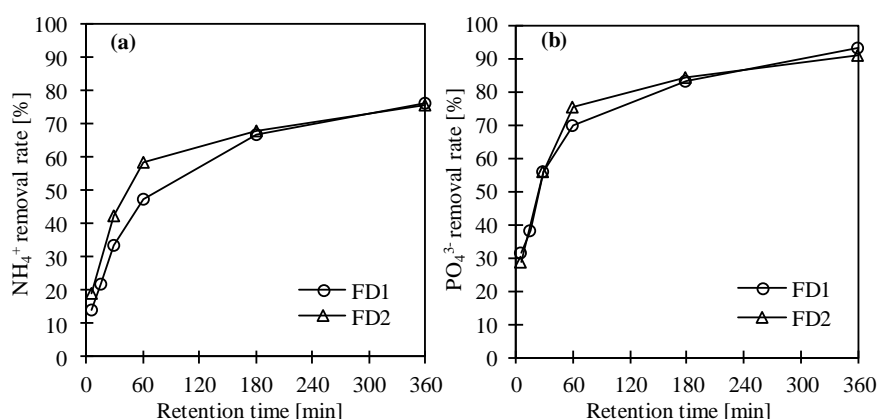


Fig. 3 Removal rates of  $\text{NH}_4^+$  (a) and  $\text{PO}_4^{3-}$  (b) calculated from the control polluted water by FD1 (circulation) in comparison with FD2 (retention).

From Fig. 3, the removal rate of  $\text{PO}_4^{3-}$  in both FD1 and FD2 was of the same order. This suggested that both circulation and retention of polluted water in the GCA filter had no influence on the removal rate of  $\text{PO}_4^{3-}$ . The removal rate of  $\text{NH}_4^+$  in both FD1 and FD2 was also of the same order when the retention time was longer than 180 min. Notably, the removal rate was different when the retention time was shorter than 180 min. A larger difference was found at the retention time of 60 min. The removal rate of  $\text{NH}_4^+$  in FD2 was 58.5%, 1.23-fold larger than that in FD1 (47.6%). In other words, the removal rate of  $\text{NH}_4^+$  by retaining polluted water was higher than that by circulating polluted water. This means that wastewater should be retained in GCA filters to obtain a higher removal rate of  $\text{NH}_4^+$  for a retention time less than 180 min.

The reason behind the different removal rates of  $\text{NH}_4^+$  between FD1 and FD2 may be partly due to different dissolution characteristics of GCA. According to Mizumoto et al. [17], dissolution of main component of GCA (fly ash) causes an increase in the solution pH. Thus, the changes in pH with different retention times in FD1 were examined compared to those in FD2, as depicted in Fig. 4. The pH of polluted water was 7.65 (Table 1), increased to 7.68 in FD1 and 7.87 in FD2 (Fig. 4). The sample pH was higher in FD2 compared to that in FD1, which is considered to be one of the factors that causes the higher removal rate of  $\text{NH}_4^+$  in FD2. This is good agreement with the suggestion made by Boopathy et al. [18] who reported that the adsorption rate of  $\text{NH}_4^+$  on coconut shell-activated carbon increased with an increase in pH. To the best of our knowledge, a decrease in hydrogen bond and an increase in electrostatic repulsion at a higher pH may contribute to the higher removal rate of  $\text{NH}_4^+$  in FD2. As a material surface rich with hydrogen ions owing to hydrogen bond, other ions are difficult to adsorb onto the material surface. Moreover, the material surface becomes more negatively charged at a higher pH. This makes ions adsorb onto the material surface easily.

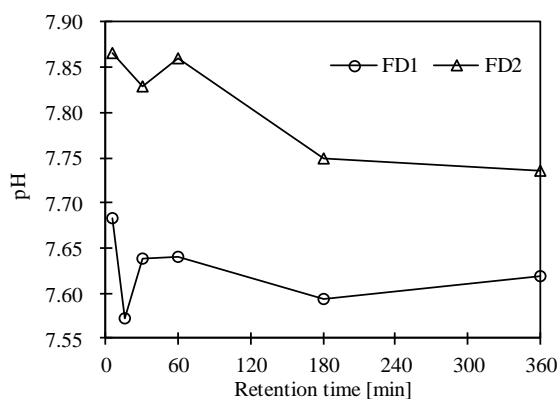


Fig. 4 Changes in pH at different retention time in FD1 in comparison with those in FD2.

### B. Absorbed Quantities of Nutrient Salts by GCA as a Function of the Concentrations of Nutrient Salts

The concentration of nutrient salts present in domestic wastewater is seasonally and regionally different. Therefore, the GCA capacity in removing nutrient salts at different concentrations of nutrient salts should be examined. Fig. 5 shows the concentrations of  $\text{NH}_4^+$  and  $\text{PO}_4^{3-}$  in the control polluted water against the absorbed quantities of  $\text{NH}_4^+$  and  $\text{PO}_4^{3-}$ . The absorbed quantity was calculated based on the concentration of nutrient salts present in the control polluted water. In this study, the absorbed quantity was united as  $\text{mg/cm}^3\text{-GCA}$  because micropores present in GCA also involve in the removal of nutrient salts. Thus, using GCA volume was thought to be better than using GCA mass. As expected, the absorbed quantities of  $\text{NH}_4^+$  and  $\text{PO}_4^{3-}$  depended strongly ( $R^2 > 0.90$ ) on the concentrations of  $\text{NH}_4^+$  and  $\text{PO}_4^{3-}$  present in the polluted water. The absorbed quantity of  $\text{NH}_4^+$  increased from 0.018 to 0.067  $\text{mg/cm}^3\text{-GCA}$  with an increase in the  $\text{NH}_4^+$  concentration from 21.5 to 57.0  $\text{mg/L}$ . Similarly, the absorbed quantity of  $\text{PO}_4^{3-}$  increased from 0.011 to 0.029  $\text{mg/cm}^3\text{-GCA}$  with an increase in the  $\text{PO}_4^{3-}$  concentration from 10.0 to 25.3  $\text{mg/L}$ . A higher adsorption of either  $\text{NH}_4^+$  or  $\text{PO}_4^{3-}$  was obtained with increasing either the  $\text{NH}_4^+$  or  $\text{PO}_4^{3-}$  concentration.

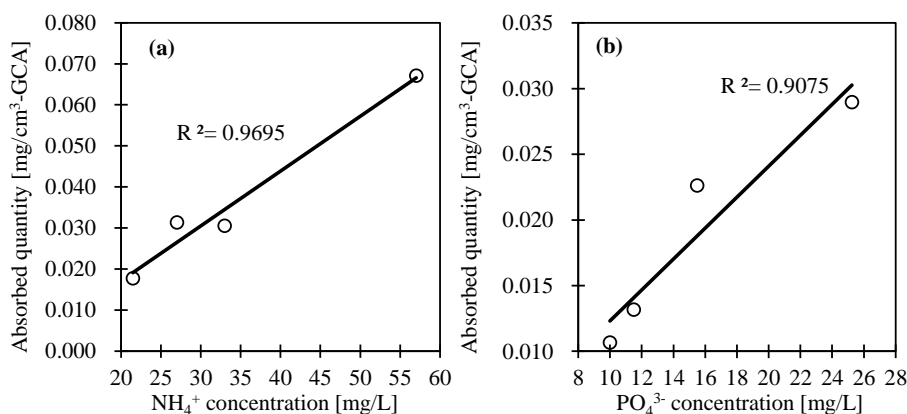


Fig. 5 Absorbed quantities of  $\text{NH}_4^+$  (a) and  $\text{PO}_4^{3-}$  (b) with regard to the concentrations of nutrient salts.

It can be understood from our results that the removal of nutrient salts by GCA does not occur due to released ions from GCA alone. As the absorbed quantity of nutrient salts depended on the concentrations of nutrient salts, it is considered that the removal of nutrient salts also occurs due to diffusion of nutrient salts into the micropores of GCA. Therefore, filter design and operation should be carefully considered for obtaining a higher removal rate of nutrient salts when using GCA.

### C. Design of GCA Filter and its Operation for a Higher Removal Rate of Nutrient Salts

From Fig. 3, the removal rate of nutrient salts decreased temporally. For instance, the removal rate of  $\text{PO}_4^{3-}$  was approximately 70-75% for the retention time of 60 min, decreased to 9-14% between the retention times of 60 min and 180 min. In wastewater treatments, the retention time should be shortened as much as possible. Here, the retention time of 90 min was chosen to examine design of the GCA filter and its operation for removing nutrient salts effectively.

As noted earlier, the absorbed quantity of nutrient salts by GCA depended on the concentration of nutrient salts present in polluted water (Fig. 5). Thus, the retaining polluted water in a GCA filter before discharging to another GCA filter for another retention (FD3-2, Case 13 in Table 2) may remove nutrient salts effectively.

Figure 6 shows the absorbed quantities of nutrient salts in FD3-1 in comparison with those in FD3-2 (two parts of FD3-1, Fig. 2). When the polluted water was retained in FD3-1 for 180 min, the absorbed quantities of  $\text{NH}_4^+$  and  $\text{PO}_4^{3-}$  were 0.018  $\text{mg/cm}^3\text{-GCA}$  and 0.011  $\text{mg/cm}^3\text{-GCA}$ , respectively.

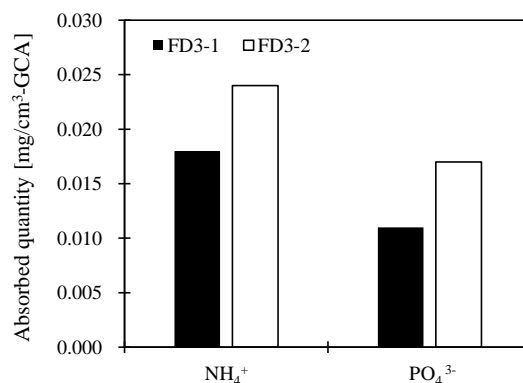


Fig. 6 Absorbed quantities of nutrient salts in FD3-1 in comparison with those in FD3-2 (two parts of FD3-1).

Interestingly, when the polluted water was retained in the first part of FD3-2 for 90 min and then discharged to the second part for 90 min-retention, the absorbed quantities of  $\text{NH}_4^+$  and  $\text{PO}_4^{3-}$  were  $0.024 \text{ mg/cm}^3\text{-GCA}$  and  $0.017 \text{ mg/cm}^3\text{-GCA}$ , respectively. As expected, a higher removal capacity was obtained owing to our proposed filter design and operation (FD3-2). 1.33-fold increase in  $\text{NH}_4^+$  removal and 1.55-fold increase in  $\text{PO}_4^{3-}$  removal were obtained. It can be said from these results that FD3-2 and its operation may be an ideal method of reducing the concentration of nutrient salts present in wastewater effectively.

#### IV. CONCLUSIONS

This study aimed to examine the potential of using GCA as a filter material in wastewater treatments, particularly, the removal of nutrient salts. Laboratory experiments were conducted to determine the removal rate of nutrient salts from polluted water by GCA filters under circulation and retention conditions of the polluted water. The removal rate of  $\text{PO}_4^{3-}$  by circulating polluted water was of the same order as that by retaining polluted water in GCA filters. However, the removal rate of  $\text{NH}_4^+$  by retaining polluted water was 1.23-fold larger compared to that by circulating polluted water for the retention time of 60 min. A higher pH was considered to contribute to a higher removal rate during the retention of polluted water compared to the circulation of polluted water. Furthermore, the absorbed quantity of nutrient salts by GCA became higher with increasing the concentration of nutrient salts. Based on these results, design of GCA filter and its operation were proposed. By retaining polluted water in a GCA filter before discharging to another GCA filter for another retention, 1.33-fold increases in  $\text{NH}_4^+$  removal and 1.55-fold increases in  $\text{PO}_4^{3-}$  removal were obtained. This suggested that nutrient salts can be effectively removed with regard to our proposed filter design and its operation.

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