Temperature effect on diffusion and non-linear adsorption of ammonium through geosynthetic clay liner and in-situ clayey soil in Hanoi, Vietnam

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ABSTRACT

High concentration of ammonium was detected in groundwater in southern Hanoi, Vietnam while municipal solid waste (MSW) landfills were known to generate large amounts of NH₄⁺. Thus, bottom barrier with well performance should be required in Hanoi MSW landfills to minimize NH₄⁺ migration. Hanoi is expected to experience temperature increase, which enables to reduce hydraulic-barrier performance. Hence, study on temperature effect on NH₄⁺ adsorption and diffusion through landfill barriers plays a key role in prevention of NH₄⁺ contaminated aquifers. The objective is to evaluate effect of three kinds of temperature (20°C, 35°C and 50°C) on NH₄⁺ adsorption and diffusion through GCL and in-situ clayey soil sampled in Hanoi (HN clay). The results show that GCL possesses higher non-linear partitioning coefficient (K_d*) and lower diffusion coefficient (D_e) than HN clay in all cases of temperature. Both GCL and HN clay experienced an increase of K_d* and D_e in the temperature range of 20°C and 35°C. When temperature rises to 50°C, K_d* tends to decrease but D_e keep increasing. Hence, the study indicates a more dominant effect of diffusion coefficient than non-linear partitioning coefficient in promotion of ammonium mass transport through HN clay.

Keywords: GCL, HN clay, non-linear adsorption, diffusion, temperature, ammonium.

1 INTRODUCTION

Vietnam is one of the most affected developing countries by climate change. Hanoi, capital of Vietnam currently possesses a considerable increment of hot summer days (> 35°C) and even increase in air temperature up to 40-41°C (Ho et al., 2011, Kubota et al., 2017). Furthermore, MSW in Hanoi were detected to contain high content of organic waste, at 71% in 2011 (JICA, 2011), hence facilitating to ammonium generation. Thus, Hanoi landfills becomes prone for high ammonium and elevated temperature. Nam Son landfill, one of the largest sanitary landfills in Hanoi city, Vietnam was recorded to contain substantial concentration of ammonium, around 1270 (±38) mg/L in July 2016 (Son et al., 2017). Meanwhile, southern of Hanoi is facing with noticeable concentration of ammonium in groundwater (Norrman et al., 2015). It therefore becomes significant for Hanoi city to offer a landfill constituted with well-performed bottom liners to protect groundwater from NH₄⁺ contamination.

GCL and clayey soil have been widely used as bottom liners of MSW landfills in recent years. These liners are commonly rich in clayey materials due to their fine grain size, micro-pores, and leading to low permeability (Du et al., 2004). However, clayey materials are considerably sensitive to not only chemical solution as NH₄⁺ but temperature change due to climate variation and waste degradation (Rowe, 2018), which are representative for current and future Hanoi situation. Therefore, evaluation of combined effects of ammonium solution and temperature change on hydraulic transport parameters of NH₄⁺ through GCL and HN clay become a primary task.

Along with permeability, diffusion is considered as a dominating process in the contaminant migration through landfill liners. Several researchers evaluated effects of temperature ranging from 6°C to 50°C on diffusion coefficient of some organic compound, heavy metals, and other cations for clayey materials such as BTEX in GCL (Rowe et al., 2005), Zn, Cd in natural clays (Do and Lee, 2006), Cr⁶⁺ in clayey soil (Yang et al., 2011), K⁺ in kaolin clay (Mon et al., 2016). However, there is so far few studies on diffusion of ammonium for GCL and clayey soil exposed to temperature change.

In addition, contaminant migration through bottom barriers is likely to be minimized due to retardation mechanism as adsorption. Many studies have
examined ammonium adsorption process onto clayey soils (Baker et al., 2010); bentonite (Pivato and Raga, 2006; Eturki et al., 2012), and GCL including bentonite and geotextile (De Oliveira Pinto et al., 2018) under effects of pH, initial bentonite, and ratio of solid/liquid, and temperature by performing adsorption test. Thus, partitioning coefficient in different temperature attained from adsorption test was one of input parameters for diffusion coefficient deduction.

Hence, the main aim of the present study is to elucidate effects of different temperature at 20, 35, and 50°C on partitioning and diffusion coefficients of NH₄⁺ through GCL and HN clay. In particular, 20°C is considered as reference temperature in experimental room; 35°C is threshold for hot summer day; and 50°C is extreme temperature. These coefficients were determined by performing adsorption and diffusion tests at this temperature range.

2 MATERIALS AND METHODS

2.1 Materials

Materials used include GCL and Hanoi clay. The GCL includes granular Na-bentonite sandwiched between a woven cover geotextile (GT) and a non-woven carrier geotextile by needle punching. HN clay was sampled from 1 m below ground surface in an expected landfill site of southern Hanoi city, Vietnam. Two materials were applied laboratory tests to obtain some physicochemical properties shown in Table 1. A specified amount of HN clay was dried in oven 110°C for 24 hours, crushed into fine grains, and then stored in a bag as adsorbent. Two soils were taken X-Ray diffraction (XRD) to examine main minerals using Ultima IV machine belonging to Rigaku Corporation, Japan. It was found that montmorillonite and quartz belonging to smectite group are major minerals of bentonite while HN clay composes mainly chamosite.

![Fig. 1. XRD results of bentonite (a), and HN clay (b).](image-url)

and quartz characterized by chloride group (Fig. 1). Solution NH₄Cl with different concentration was utilized as contaminant fluid for adsorption and diffusion tests. These stock solutions were prepared by dissolving NH₄Cl powders (analytical reagent) in deionized (DI) water.

2.2 Methods

Adsorption test

The adsorption test at different temperature (20, 35, 50°C) was performed to determine effects of temperature change on NH₄⁺ adsorption of bentonite and HN clay. Bentonite adsorption is typical for GCL adsorption because geotextile was studied to have few ammonium adsorption (De Oliveira Pinto et al., 2018). Based on adsorption test, mass of absorbed NH₄⁺ into soils were calculated has been obtained. 2 g of dry bentonite and HN clay was filled in 50 ml plastic centrifuge tubes with 40 ml solution NH₄Cl in different concentration. The initial NH₄⁺ target concentration of 10, 50, 100, 200, 500, and 1000 mg/L was prepared. The tubes were equilibrated by shaking at a speed of 200 rpm (revolutions per minute) for at least 24 hours inside a temperature-controlled incubator and then centrifuged at 4000 rpm for 30 min. The blank samples of these concentration were also tested to examine

| Property         | Bentonite | HN clay |
|------------------|-----------|---------|
| Physical property|           |         |
| Water content (%)| 12.0      | 47.40   |
| Liquid limit (LL) (%)| -        | 82.10   |
| Plasticity index (PI) (%)| -        | 41.40   |
| Specific gravity | 2.61      | 2.68    |
| Total porosity (-) | 0.82    | 0.63    |
| Classification (UCS) | CH      | CH      |
| pH               | 8.2       | 6.2     |
| Organic content (%)| 2.47     | 3.28    |
| Bulk chemical composition (%)|        |         |
| SiO₂            | 57.1      | 54.16   |
| Al₂O₃           | 19.8      | 20.22   |
| Fe₂O₃           | 4.75      | 6.48    |
| K₂O             | 0.65      | 2.97    |
| MgO             | 2.57      | 1.22    |
| TiO₂            | 0.3       | 0.98    |
| CaO             | 1.86      | 0.46    |
| Na₂O            | 2.36      | 0.34    |

Table 1. Physicochemical properties of bentonite and HN clay.

Table 2. Mathematical equations of isotherm models used (Rowe et al., 2004).

| Isotherm | Isotherm form | Partitioning coefficient |
|----------|---------------|--------------------------|
| Linear   | \( S = K_f c_e \) | (1)                      |
| Non-linear |               |                          |
| Langmuir | \( S = \frac{S_m b c_e}{1 + b c_e} \) | (2) \( K_d^* = \frac{S_m b}{1 + b c^*} \) | (3) |
| Freundlich | \( S = K_f c^e \) | (4) \( K_d^* = K_f c^{e^*(1-\epsilon)} \) | (5) |

\( S_m \) : maximum adsorption capacities, \( b \) : Langmuir isotherm constant, \( K_f \) : Freundlich isotherm constant, \( c^* \) : adsorption intensity, \( c^* \) : the estimated concentration at the point and time of interest, \( K_d^* \), \( K_d^* \): linear and non-linear partitioning coefficient, respectively.
evaporation of ammonium. The samples were taken 1.0 ml from the supernatant by using pipette. The concentration of NH$_4^+$ was analyzed using UV-VIS spectrophotometer belonging to Kumamoto University.

For modelling adsorption isotherm, concentration of NH$_4^+$ at equilibrium and its mass removed from solution were used. Of which, the NH$_4^+$ uptake by bentonite and HN clay can be expressed as:

$$S = \frac{(C_0 - C_e)V}{m}$$  \hspace{1cm} (6)

where $S$ is mass of contaminant removed from solution (mg/kg); $C_0$ and $C_e$ are initial and equilibrium concentration of adsorbate, respectively (mg/L); $V$ is volume of solution (L); $m$ is mass of adsorbent (kg).

The equilibrium concentration of NH$_4^+$ and mass of NH$_4^+$ removed from solution can be expressed with isotherm models including linear or non-liner isoplets (Langmuir and Freundlich). From these isoplets, partitioning coefficient defined as ratio of quantity of NH$_4^+$ adsorbed per unit mass of bentonite/ HN clay to quantity of NH$_4^+$ remained in solution at equilibrium was calculated by mathematical equations in Table 2.

### Diffusion test

Figure 2 indicates the schematic of diffusion cell used in specified volume diffusion test. The diffusion cells are made from vinyl chloride materials to avoid ammonium evaporation. The cell of 90 x 68 mm in height and diameter composes of a 10-15 mm thickness of middle-pre-consolidated soil/GCL between 50-mm-thick upper (source) and 30 mm-thick lower (receptor) rings. In case of GCL, pre-hydration process from the bottom of the sample for swelling needed being completed before diffusion test initiation. The prepared solutions of 1000 mg/L NH$_4^+$ (750 mg/L NH$_4^+$ - N) and DI water were filled in source and receptors reservoirs, inside a temperature-controlled incubator at different temperature including 20, 35, and 50 oC. Like adsorption test, the blank solution of 1000 mg/L NH$_4^+$ without soil samples was tested in the same condition to evaluate the evaporation of ammonium. 1.0 ml samples of solution at source and receptor was taken every two days and replaced with equal volume of DI water. NH$_4^+$ concentration analysis procedure for these samples is same as that of adsorption test.

In addition, theoretical model used in this paper was originated from one-dimensional finite-layer method inferred from running the program POLLUTE (Rowe and Booker, 1998). The one-dimensional partitioning and diffusion equation can be written as below equation with $K_d/K_d^*$ and dry density ($\rho_d$) as inputs.

$$\frac{\partial c}{\partial t} = \frac{D_e}{(1 + \frac{Pd.K_d}{n})} \frac{\partial^2 c}{\partial z^2}$$  \hspace{1cm} (7)

where $c$ is a concentration at depth $z$ and time $t$ [ML$^{-3}$]; $n$ = porosity [-] .

POLLUTE v7 (version 7) using various deduced $D_e$ generates the output charts of concentration of chemical solution vs time in source and receptor reservoir. The fitness between these data and the above experimental data was evaluated based on both visual fit and square summation error (SSE) calculated by equation (8) to determine the reasonable value of $D_e$.

$$SSE = \sum \{x_i - x'_i\}^2$$  \hspace{1cm} (8)

where $\{x_i\}$ is the experimental data series; $\{x'_i\}$ is the POLLUTE data series.

### 3 RESULTS AND DISCUSSION

#### 3.1 Effects of temperature change on ammonium adsorption

Figure 3 illustrates that NH$_4^+$ exhibits a non-linear adsorption isotherm over the range of concentrations tested with bentonite and HN clay. This non-linear adsorption data was the best fit to Langmuir sorption model for both bentonite and HN clay. Langmuir parameters including adsorption capacity $S_m$ (mg/kg), Langmuir constant $b$, and $R^2$ are shown in Table 3. A non-linear partition coefficient $K_d^*$ at concentration of 750 mg/L NH$_4^+$- N calculated from equation (3) was also shown. It can be noted that values of $K_d^*$ for both bentonite and HN clay saw a slight increase from 20$^\circ$C to 35$^\circ$C and then decreased in the range of 35$^\circ$C and 50$^\circ$C. This result is consistent with the conclusions of Baker et al. (2010); and Chen et al. (2018) that removal efficiency of NH$_4^+$ ion rose with the increase of temperature when temperature is lower than room temperature (about 25-35°C) but in this study it decreased with the increase of temperature when temperature is higher than room temperature. Eturki et al., (2012) investigated NH$_4^+$ removal capacity in the temperature range of 20-to-40°C. It was found that the amount of ammonium removed onto solid increased with temperature increasing. In essence, an increase in temperature clearly enhances rate of chemical

![Fig. 2. Schematic of diffusion cell.](image-url)
Table 3. Langmuir parameters of bentonite and HN clay.

| Parameters | Temperature (°C) |
|------------|------------------|
|            | Unit 20 35 50    |
| Bentonite  |                  |
| $S_m$      | mg/kg 10638 13793 16667 |
| $b \times 10^{-3}$ | L/mg 1.9 2.1 1.0 |
| $K_d^*$    | mL/g $1.5 \pm 0.1$ $1.9 \pm 0.0$ $1.6 \pm 0.2$ |
| $R^2$      | - 0.93 0.99 0.98 |

$K_d^*$: partitioning coefficient from non-linear isopleth at estimated concentration of 750 mg/L $NH_4^+$

Fig. 3. Isotherm of ammonium adsorption onto bentonite and HN clay at different temperature.

adsorption reaction between adsorbate and adsorbent as Arrhenius equation (Clark 1996). However, subjecting to high temperature of the solution at 50°C in this study, there may be tendency for $NH_4^+$ ion to desorb from the solid phase to the bulk phase once equilibrium state was reached (Karadag et al., 2006; Baker et al., 2010). In addition, $K_d^*$ of bentonite was found to be 5 times higher than HN clay. Hence, smectite group in bentonite with higher surface area and CEC contributes to a much more significant adsorption than chloride group of HN clay.

3.2 Effects of temperature change on ammonium diffusion

Figures 4 and 5 indicate the normalized concentration ($C/C_0$) of $NH_4^+$ in the source and receptor of the GCLs and HN clays tested at 20, 35 and 50°C respectively from experiments (plots) and theoretical modelling (lines). In term of GCLs, the theoretical modelling is performed through both 1 layer of the entire GCL and 3 layers including cover geotextile (GT) intruded by bentonite, bentonite and carrier geotextile intruded by bentonite. It is assumed that bentonite is uniformly distributed in all layers. The porosity of each layer of GCL and the porosity of the entire GCL was determined based on the definition of the void ratio of the GCL, referred from Rowe et al. (2005). Particularly, the total porosity of cover GT, bentonite, and carrier GT at 20°C, 35°C, and 50°C is (0.67; 0.85; 0.65), (0.66; 0.84; 0.64), (0.65; 0.83; 0.63), respectively. The total porosity of the entire GCL at 20°C, 35°C, and 50°C is 0.82, 0.81, and 0.81, respectively. It can be noted that there is a negligible change in porosity when temperature changes from 20°C to 50°C. From diffusion curves of the GCL in Fig. 4, the better fit at the initial stage between measured and predicted concentration at source is observed for the case of 3 layers than 1 layer. Furthermore, it can be seen in Fig. 4 and 5 that ammonium concentration at receptor increases in the increment of temperature and that at source decreases when temperature rises for both GCL and HN clay. And diffusive curves of HN clay (Fig. 5) appeared to be steeper than those of HN clay (Fig. 4). Basically, the equilibrium normalized concentration between the source and receptor of both GCL and HN clay was obtained after 14-day diffusion at 50°C and took longer time for lower temperature. Indeed, these observations suggest faster diffusive migration of $NH_4^+$ at higher temperature. By modelling predicted $NH_4^+$ concentration, the deduced $D_e$ was back-calculated by iterating $D_e$ values until the concentration profile fit the experimental data well with the smallest SSE. Table 4 shows total diffusion

Fig. 4. Normalized ammonium concentration ($C/C_0$) at source and receptor during elapsed time of GCL.
coefficient of NH$_4^+$ at different temperature for GCL and HN clay and their SSE values. The diffusion coefficients of the entire GCL (1 layer) and 3 layers of geotextiles and bentonite are relatively similar. The total diffusion coefficient increases in the range of 6.6-6.9 x 10^{-10} and 7.2 x 10^{-10} m$^2$/s for GCL; 7.5 x 10^{-10} and 8.5 x 10^{-10} m$^2$/s for HN clay when temperature rises from 20$^\circ$C to 35$^\circ$C. In the temperature range of 35$^\circ$C and 50$^\circ$C, diffusion coefficient keeps rising until 8.0 x 10^{-10} m$^2$/s for GCL and 1.1 x 10^{-9} m$^2$/s for HN clay. The bentonite composes of a different range of pore sizes including aggregates (inter-aggregate), particles (inter-particle), and layers (interlayer) (Acikel et al., 2018). According to Villar and Lloret (2004), the increase in temperature transfers water absorbed in inter-particle pores into free water in inter-aggregate pores. Whereas, the ammonium ions have a tendency to diffuse in free water in inter-aggregate pores. Probably, the temperature increase diffusion coefficient of the GCL with ammonium owing to not only the increase in mobility of ammonium but also larger inter-aggregate pores of bentonite.

3.3 Dominant parameter due to temperature change

Figure 6 summarizes temperature change effects on trend of non-linear partitioning and diffusion coefficients. The trend of both coefficients for GCL and HN clay is mostly similar. Increment rate of $D_e$ for HN clay is faster than that for bentonite. Both partitioning and diffusion coefficients contribute to increase of NH$_4^+$ diffusive mass transport, therefore, a comparison between $K_d$ and $D_e$ was performed in order to determine the dominant parameter controlling diffusion. Assuming that theoretical diffusive curves mixing $K_d$ and $D_e$ between 20$^\circ$C and 35$^\circ$C as well as 35$^\circ$C and 50$^\circ$C including (b) $K_d$ & $D_e$, (c) $K_d$ & $D_e$.

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Fig. 6. Trend of partitioning coefficient and diffusion coefficient in temperature change of GCL (a) and HN clay (b).

Fig. 7. Theoretically diffusive curves obtained from three combinations of $D_e$ and $K_d$ for HN clay in cases of 20$^\circ$C & 35$^\circ$C (I) and 35$^\circ$C & 50$^\circ$C (II).
The adsorption and diffusion tests were performed to evaluate adsorption and diffusion parameters for ammonium of GCLs and HN clays at three temperature of 20°C, 35°C and 50°C. GCL finds higher partitioning coefficient and lower diffusion coefficient than HN clay in all cases of temperature. For both GCL and HN clay, partitioning coefficient rises from 20 to 35°C and drops from 35 to 50°C but diffusion coefficient keeps increasing. Diffusion coefficient dominates over partitioning coefficient in enhancing NH₄⁺ diffusive transport through HN clay when temperature increases. The results become useful reference data for suitable material selection in barrier design for Hanoi landfills suffering from temperature change.

ACKNOWLEDGEMENTS

The authors would like to express thanks for the financial support of Ministry of Education, Culture, Sport, Science and Technology (MEXT) – Japan. We also highly appreciate ammonium analysis supports of Associate Prof. Takehide Hama of Kumamoto and preparation of diffusion cell by Mr. Toru Yoshinaga, who is a technical staff of Kumamoto University.

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