

Effect of pH on arsenic removal performance of hydrotalcite-coated geosynthetic sorption sheet

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ABSTRACT

In recent years, attenuation layer method has been considered an effective countermeasure for excavated soils and rocks containing toxic elements like arsenic (As). Non-woven geosynthetic sorption sheets coated with hydrotalcite on one side can be used in the attenuation layer method as a promising sorption material in permanent geostructures and for temporary storage. The sorption sheets used in the attenuation layer can be exposed directly to leachate from the excavated soil and rocks. The pH of leachate differs by site and time due to large spatial variations in chemistry and mineralogy and oxidation with time. In this study, the As removal efficiency of the geosynthetic sorption sheet was investigated to ensure its performance with the main focus on solution pH. Batch sorption experiments were conducted to evaluate the As removal from aqueous solution with various contact times (5 min–24 h) and initial As concentrations (0.1–20 mg/L) against different solution pH (3–9). Results revealed that the geosynthetic sorption sheet is efficient in As sorption, but As(V) is more efficiently removed than As(III). The As(III) sorption was strongly affected by solution pH. As(III) sorption mass decreased by 15% as the solution pH decreased from 9 to 3. In comparison, the sorption of As(V) was pH-independent, and its sorption mass was almost identical in the pH range of 3–9. The partial dissolution of hydrotalcite was confirmed by measuring the concentration of precursor metals (Mg, Al) in hydrotalcite during the sorption process.

Keywords: Arsenic, Excavated soils and rocks, Geosynthetics, Sorption, Hydrotalcite, Dissolution

1 INTRODUCTION

In the construction process of tunnels, large amounts of excavated soils and rocks are generated, which is a severe problem for environmental and social security (Katsumi, 2015; Tu et al., 2023). In Japan, for instance, more than 10,000 tunnels longer than 5,000 kilometers have been built, respecting more than 280 million m³ of excavated material annually produced, according to the latest data released from MLIT (MLIT 2018). Therefore, how to treat these excavated solid materials has become an essential consideration in tunnel construction projects. (Bai et al., 2022). With the increasing awareness of sustainable development, the recycling of excavated soils and rocks as geomaterial has been highly encouraged during the last decade (Brandely et al., 2022; Kato et al., 2021; Magnusson et al., 2019). However, the corresponding environmental and health considerations have increasingly become a key factor affecting the recycling of excavated soils and rocks (Haas et al., 2021; Gathuka et al., 2021).

Arsenic (As) is a nonmetallic element that occurs in trace amounts and is widely distributed in natural sedimentary soils and rocks (Li et al., 2020; Ho et al., 2021; Gathuka et al., 2022). Long-term exposure to arsenic pollution would seriously affect human health and increase the risk of developing cancer (Yadav et al., 2017; Wei et al., 2011). It is known that arsenic concentration in Japan maintains at a relatively higher level compared with the world average, primarily because Japan is located in the convergence part of the Eurasian Plate and the Pacific Plate with frequent crustal activity (volcanism and the related activities) (Tabelin et al., 2012; Jha and Tripathi, 2021). Recently several studies have indicated that the leaching concentration of As from excavated soils and rocks sometimes exceeded the Japanese environmental standard of 10 µg/L for arsenic, which has aroused widespread concern in Japan (Li et al., 2016, 2020). Thus, it is important to establish effective methods and frameworks to deal with such excavated soils and rocks.

The sorption layer method is one of the effective methods for such naturally contaminated soils and rocks, realizing the attenuation of pollutants from leachate by sorption layer (Mo et al., 2021). Although many types of sorption material have been developed, a hydrotalcite-coated geosynthetic sorption sheet is used in this study. Geosynthetic products have been widely applied in geo-environmental engineering over recent years (Weerasinghe et al., 2020). The hydrotalcite-coated geosynthetic sorption sheet is a composite geosynthetic material with one face that consists of solidified hydrotalcite attached to the non-woven geotextile. This material could be applied in various field practices (embankment constructions or temporary storage facilities), depending on the reusing mode of excavated soils and rocks (Kinoshita et al., 2021). The outstanding mechanical properties of geosynthetic products can prevent them from being damaged during the period of service (Ding et al., 2021). The hydrotalcite contained in the coated face plays an essential role in attenuating contamination. Numerous investigations have indicated that hydrotalcite is a promising sorbent with a high sorption performance for arsenic (Sanchez-Cantu et al., 2016; Türk and Alp, 2014; Chetia et al., 2012).

Arsenic in water typically exists in trivalent [As(III), arsenite] and pentavalent [As(V), arsenate] forms, which are primarily affected by the solution pH, thereby affecting the sorption process (Dias and Fontes, 2020). The speciation distributions of As in solutions concerning pH have been well studied (Smedley and Kinniburgh, 2002). There have been several studies evaluating leaching characteristics from tunnel-excavated soils and rocks (Tabelin and Igarashi, 2009; Tabelin et al., 2014a,b). The results showed that the pH of leachate differs by site and time due to large spatial variations in chemistry and mineralogy. On the other hand, some studies also expressed concerns about the structural instability of hydrotalcite during the sorption process, suggesting that the dissolution of hydrotalcite would occur in certain pH conditions (Dias et al., 2019; Dias and Fontes, 2020; Goh et al., 2008).

The present study aimed to explore arsenic (As(III) and As(V)) sorption by a hydrotalcite-coated geosynthetic sorption sheet. A series of batch sorption experiments were conducted to evaluate its sorption performance as a function of contact time, initial concentration, and solution pH. The dissolution of hydrotalcite during the sorption process was also evaluated.

2 MATERIALS AND METHODS

2.1 Materials

The geosynthetic sorption sheet with a nominal thickness of 2.8 mm is a nonwoven product with an areal density of 450 g/m² (310 g/m² for nonwoven fabric and 90 g/m² for additives in processing), which carries a hydrotalcite coating (mass per unit area is 50 g/m²) on one side. The hydrotalcite coating was generated by an advanced coating technology that provides precise thickness control of the coating. The schematic of the geosynthetic sorption sheet is shown in Figure 1. For illustration purposes, the terms “nonwoven fabric layer” and “hydrotalcite face” will be used to distinguish the uncoated side and coated side in the following. In terms of structure, as shown in Figure 1, the geosynthetic sorption sheet presents a double-layer structure: a non-woven fabric layer composed of long polyester fiber and a hydrotalcite face. First, the porous structure of the non-woven fabric layer typically enhances the solution diffusion. Second, the hydrotalcite face ensures the reliability of sorption performance. The hydrotalcite contained in this geosynthetic sorption sheet is chloride hydrotalcite having an Mg/Al atomic ratio of 2–5.

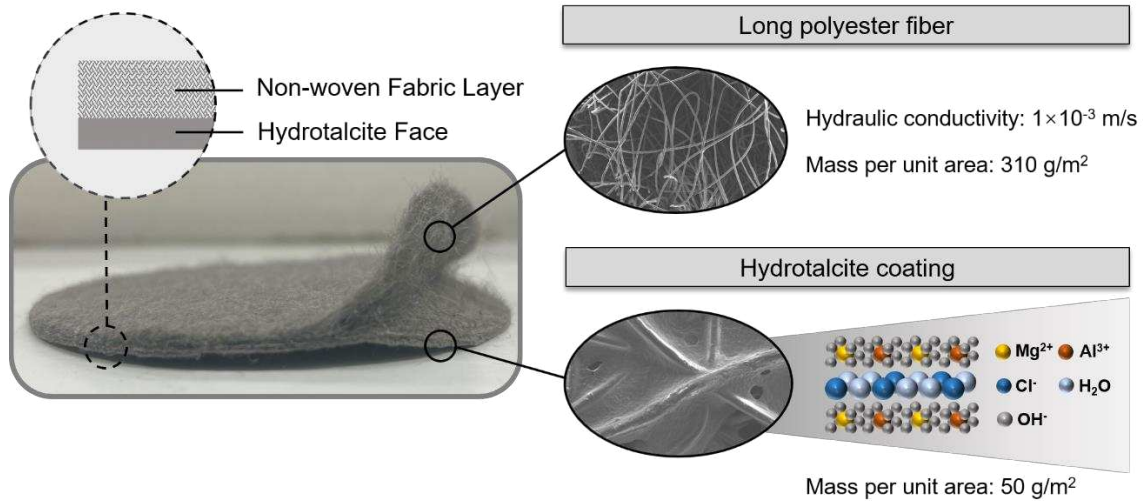


Figure 1. Schematic illustration of hydrotalcite-coated geosynthetic sorption sheet

Sodium dihydrogen arsenate (Na_2HAsO_4 , $\geq 98\%$, Aldrich) and sodium arsenite (NaAsO_2 , $\geq 90\%$, Aldrich) were used for the preparation of arsenic (As(III) and As(V)) solutions, respectively. Hydrochloric acid (HCl, 1 mol/L, Wako) and sodium hydroxide (NaOH, 1 mol/L, Wako) solutions were used for the adjustment of pH value.

2.2 Batch sorption experiments

The sorption of arsenic (As(III) and As(V)) on the geosynthetic sorption sheet as a function of time was first investigated with a fixed concentration of 0.1 mg/L for checking the equilibrium time. A pre-cut geosynthetic sorption sheet with a size of 5 cm \times 8 cm (containing 0.2 g sorbent) was added into the 250-mL sealed plastic bottle containing 200 mL arsenic (As(III) and As(V)) solution. These plastic bottles were then placed into a reciprocal oscillator (TS-10, TAITEC Co., Ltd.) running at 150 rpm and at $20 \pm 2^\circ\text{C}$. After the predetermined contact time (5 min–24 h), the solution samples were collected to analyze the As concentration after filtration using 0.45 μm -opening membrane filter. All the batch sorption experiments were conducted in triplicate, the specific steps are presented in Figure 2.

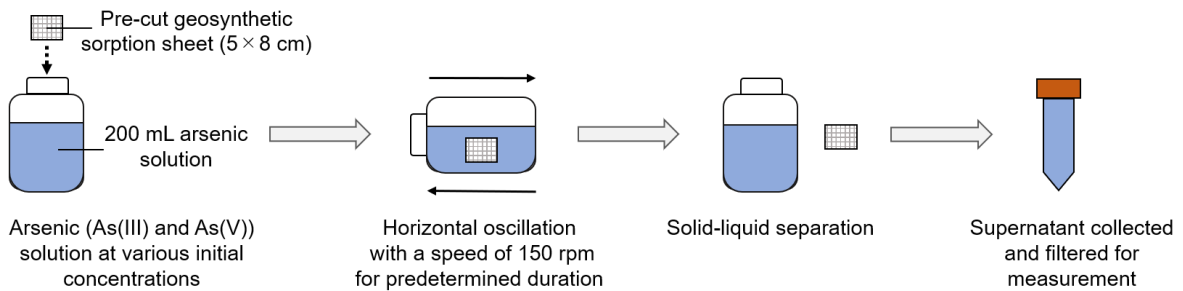


Figure 2. The schematic diagram for the procedure of the batch sorption experiments

The sorption equilibrium experiments were then carried out with arsenic (As(III) and As(V)) solution at concentrations of 0.1, 1, 10, and 20 mg/L. The contact time was determined as 24-h, confirmed by the above equilibrium time studies to be sufficient for sorption equilibrium. Once equilibrium was achieved, the suspension was filtered, and the As concentration was determined. The experimental procedures were consistent with the above-mentioned sorption tests detailed.

The sorption mass (S , mg/g) and removal ratio (R , %) were calculated using Equations (1) and (2), respectively, as follows:

$$S = (C_0 - C)V/m \quad (1)$$

$$R = (C_0 - C)/C_0 \times 100 \quad (2)$$

where C_0 represents the initial concentration of As (mg/L), C is the As concentration at any time or equilibrium (mg/L), V refers to the volume of As solution (mL), and m is the mass of the sorbent (g).

2.3 Influence of initial pH on arsenic sorption

Samples (pre-cut at a size of 5 cm × 8 cm) of geosynthetic sorption sheet were placed into separate plastic bottles and mixed with 200 mL As solutions at a fixed concentration of 20 mg/L. The initial pH of all solutions was separately adjusted to be 3, 5, 7, and 9 using HCl and NaOH solutions, the volumes of which are negligible for the whole solution. Seal bottles were placed into a reciprocal oscillator and shaken at $20 \pm 2^\circ\text{C}$ and 150 rpm for 24 h until a complete equilibrium was achieved. The suspension samples were then filtered by passing through 0.45 μm filters and kept for the quantitative detection of As concentrations. In addition, the solution pH after equilibrium was also measured.

2.4 Analytical methods

The micromorphology of geosynthetic sorption sheets was evaluated using a scanning electron microscope (SEM) (JSM-6390LVS, JEOL). The concentrations of arsenic and other major elements in solution were investigated by flame atomic adsorption spectrometry (AAS) (AA-6800, SHIMADZU) and inductively coupled plasma optical emission spectrometry (ICP-OES) (710 ICP-OES, Agilent Technologies), respectively. Evaluation of the pH variation in solution was carried out by pH meter (F-54S, HORIBA).

3 RESULTS AND DISCUSSION

3.1 Influence of contact time

The results of the removal ratio of arsenic (As(III) and As(V)) as a function of contact time are presented in Figure 3. It can be seen that the removal ratio of both As(III) and As(V) with an initial concentration of 0.1 mg/L increased with the contact time. Correspondingly, the removal ratio of As(III) and As(V) were 37% and 85% at a contact time of 5 min, respectively, increasing to 93% and 97% after a contact time of 24 h, respectively. The initial phase of rapid sorption, as shown in Figure 2, was observed during the first hour of contact, and a steady state of equilibrium was obtained at around 24 h. The removal ratio during the initial phase of the sorption process (5 min–1 h) increased from 37% to 93% for As(III) and from 85% to 97% for As(V). These results implied that the high sorption efficiency for arsenic could be achieved in a short period. This may be attributed to the more sorption sites available for arsenic on the geosynthetic sorption sheet at the beginning of the sorption process (El-Sayed, 2011). It is noteworthy that the removal ratio at equilibrium for both As(III) and As(V) reached upwards of 93%, which suggests that the geosynthetic sorption sheet has an excellent sorption performance for arsenic.

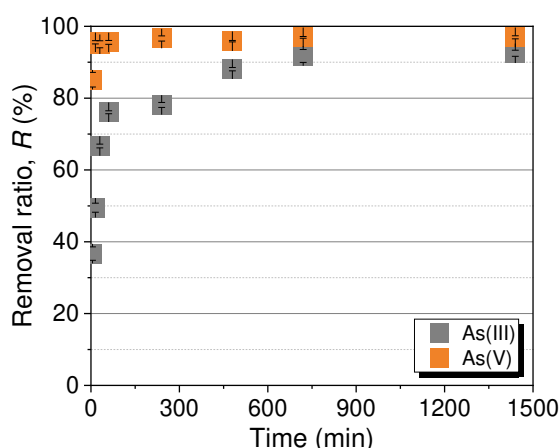


Figure 3. Effect of contact time on the removal ratio of As(III) and As(V) at a fixed concentration of 0.1 mg/L

3.2 Influence of initial concentration

The effect of initial concentration on the arsenic sorption was investigated in the range of 0.1–20 mg/L, and the results are presented in Figure 4. It was observed that the sorption mass of arsenic (As(III) and As(V)) increased with initial concentration. At any given initial concentration, the sorption mass of As(V) is higher than that of As(III). For example, after the equilibrium time of 24 h, the sorption mass of As(V) cases with initial concentrations of 0.1, 1, 10, and 20 mg/L were 0.10, 0.98, 11.05, and 19.15 mg/g, respectively. On the other hand, for the results from As(III) cases, the sorption mass was 0.10, 0.92, 6.85, and 14.54 mg/g for 0.1, 1, 10, and 20 mg/L, respectively. The results showed that As(V) is easier to be sorbed by the geosynthetic sorption sheet than As(III). The significant discrepancies in sorption performance between As(III) and As(V) can be interpreted from the different species of arsenic in aqueous solutions (Urbano et al., 2012). The monitoring data of pH during the equilibrium sorption experiments showed that the solution pH values during the sorption process kept stable within the range of 8.4–9.0. Within this range, according to the speciation distributions of As in solutions concerning pH, As(III) exists mainly as the nonionic form (H_3AsO_3), while As(V) is dominant in the form of the negatively charged species (HAsO_4^{2-} and H_2AsO_4^-) (Smedley and Kinniburgh, 2002; Yang et al., 2006). The sorbent in the geosynthetic sorption sheet is hydrotalcite, the sorption mechanism of which essentially involves the interlayer anion exchange (Nakamura et al., 2021). Therefore, it would contribute to the sorption of negatively charged As(V) species onto the geosynthetic sorption sheet.

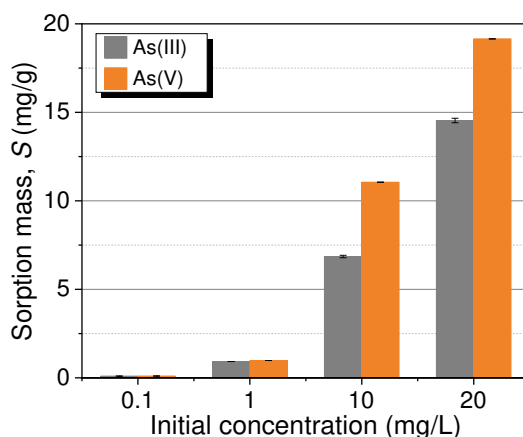


Figure 4. Effect of initial concentration on the equilibrium sorption mass of As(III) and As(V)

3.3 Influence of initial pH

The influence of initial pH on the arsenic (As(III) and As(V)) sorption by the geosynthetic sorption sheet with an initial concentration of 20 mg/L is presented in Figure 5. As shown, for As(V) cases, there is a weak correlation between the sorption mass of As(V) and the solution pH. The sorption results showed that the sorption mass of As(V) was almost identical in the pH range of 3–9 (20.7–20.8 mg/g). However, the sorption mass of As(III) increased as the initial pH of the solution augmented. It can be observed that the sorption mass of As(III) increased from 12.5 to 14.7 mg/g with the increasing solution pH from 3 to 9, leading to around a 17.6% increase in sorption mass. At an extreme pH of 3, almost no As(III) exists with negatively charged ionic forms, so it is difficult to achieve As(III) sorption through anion exchange mechanism (Yang et al., 2005). On the contrary, the higher sorption of As(III) at high-pH solutions could be explained by the presence of negatively charged species (H_2AsO_3^-).

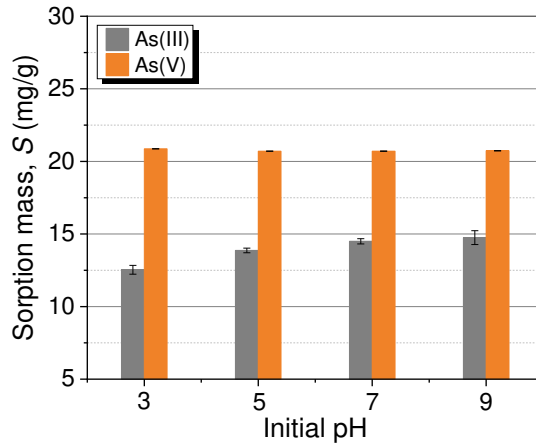


Figure 5. Effect of pH on arsenic sorption

Moreover, the pH values of solutions at equilibrium were also evaluated, as shown in Figure 6 (left). The buffering action of hydrotalcite was observed during the sorption process, no matter for the acid solution or the base solution, maintaining the equilibrium pH values of the final solutions in the range of 8.49–8.90. The increase in the equilibrium pH value of final solutions for the cases at the lower pH range may be due to the partial dissolution of the sorbent in the low-pH solutions (Yang et al., 2005). This point can be proved by the results detected from ICP-OES that dissolved precursor metals (Mg, Al) in hydrotalcite were found in the final solution. Figure 6 (right) shows the concentration of Mg^{2+} and Al^{3+} in the final solutions with different initial pH after an equilibrium time of 24 h. In the pH range of 3.0–9.0, the dissolution of Al^{3+} seems to be pH-independent, and its concentration kept stable around 3.87 mg/L. In comparison, the concentration of Mg^{2+} in solutions was strongly related to pH values. The results indicated that the concentration of Mg^{2+} at pH 3 is more than 1.5 times that at pH 9. Moreover, the dissolution of Mg^{2+} is much higher than that of Al^{3+} , mainly due to the Mg/Al atomic ratio (ratio of Mg/Al = 2–5, see section 2.1) of the hydrotalcite contained in the geosynthetic sorption sheet.

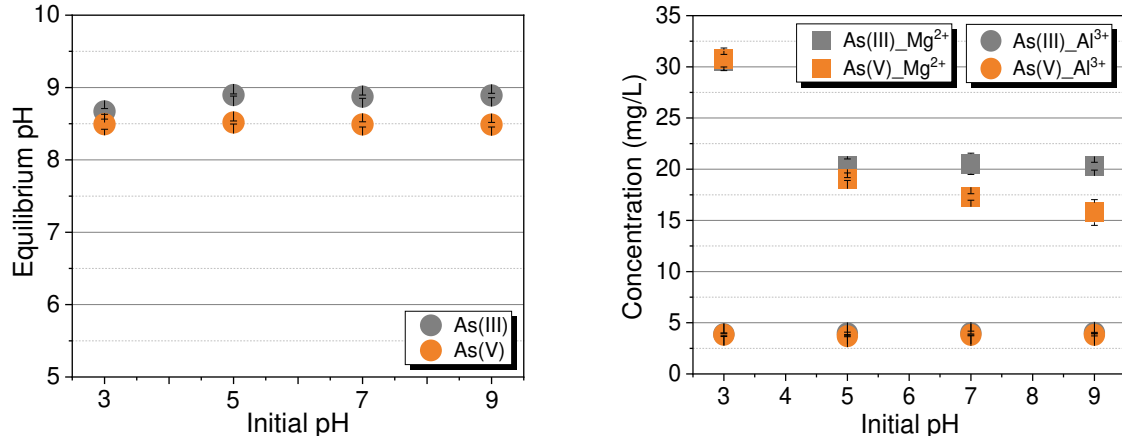


Figure 6. Relation curves between initial and equilibrium pH (left). Mg and Al concentrations in the solution with different initial pH (right)

4 CONCLUSIONS

The hydrotalcite-coated geosynthetic sorption sheet used in this work showed an excellent sorption performance for arsenic (As(III) and As(V)), and the removal ratio was more than 90%. Compared with As(III), it had a higher sorption mass for As(V) at any given initial concentration. Since the primary sorption mechanism of hydrotalcite is the interlayer anion exchange, which could contribute to the sorption of negatively charged As(V) species onto the geosynthetic sorption sheet. Furthermore, the As(III) sorption is strongly related to the initial pH in solutions, decreasing by 15% in As(III) sorption mass as the solution pH decreased from 3 to 9. In the extreme acid pH 3 almost no As(III) exists with negatively charged ionic forms. However, with the increasing solution pH, the negatively charged species ($H_2AsO_3^-$) gradually increases, which promotes the anion exchange mechanism. On the contrary, the correlation between the As(V) sorption and the solution pH is not significant. Finally, the

chemical analysis results of the final solutions confirmed the partial dissolution of hydrotalcite during the sorption process, which buffered the equilibrium solution pH, regardless of the acid or base conditions.

In practical applications, the geosynthetic sorption sheets used as a sorption layer will inevitably be exposed to leachate from the excavated soil and rocks. Fluctuations in chemistry and mineralogy can extensively alter the leaching characteristics, by which the attenuation of contamination was significantly affected. The present study investigated the As removal efficiency of this geosynthetic sorption sheet through batch sorption experiments, with the main concern on the effect of different solution pH on the sorption performance. It is also notable that the complexity in leaching characteristics of leachate is not only reflected in its variable pH values but also in the presence of various coexisting ions (such as SO_4^{2-} , and HCO_3^-) (Tabelin et al., 2014a). It has been reported that these common oxyanions potentially influence arsenic oxyanions sorption by hydrotalcite due to the competition for the available sorption sites (Goh and Lim, 2010). Therefore, further experimental studies are required to explore the effect of various co-existing ions on the arsenic removal performance of the hydrotalcite-coated geosynthetic sorption sheet.

5 ACKNOWLEDGEMENTS

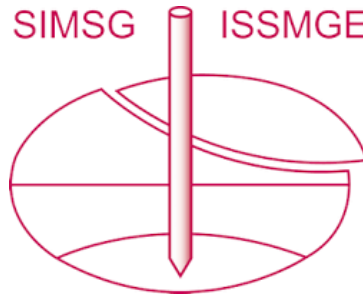
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