Control of pH of retained water in the coastal waste disposal site

Ramrav Hem1,*, Tomohiro Yamasaki1, and Hiroko Sumida1

1Geoenvironment Laboratory, Technical Research Institute, Toyo Construction Co., Ltd., 1-25-1, Naruohama, Nishinomiya City, Hyogo Prefecture, 663-8142, Japan

Abstract. After landfilling of wastes is completed, the stabilization of landfilled ground requires much time and cost. Therefore, this study aimed to control the pH of retained water in the coastal waste disposal sites during landfilling process, by conducting field surveys and laboratory experiments. In field surveys, we investigated the changes of retained water quality such as pH, salinity, and dissolved oxygen. The results show the pH of retained water has risen to about 10 when the volume of landfilled wastes reached about 25% of landfill capacity. In lowering the pH, we considered a low-cost method by pumping seawater from the adjacent sea into the landfill. The mechanism in this method is that, H+ dissociated from HCO3- in the fresh seawater react with OH- eluted from wastes would result in pH decrease. The laboratory experiments were conducted to verify the effect on pH change by adding fresh seawater to alkalized seawater. As a result, the effect of injecting fresh seawater into alkalized seawater with pH higher than 9 was confirmed. Therefore, this treatment method is suggested to enable the disposal sites to be used promptly after landfilling is completed, by adding fresh seawater to purify the retained water and waste at low cost during landfilling process.

1 Introduction

In Japan, especially in the metropolitan areas with narrow in-land spaces, waste disposal facilities are built at the coastal and sea areas. It is known that seawater has pH buffering capacity [1]. However, at some disposal sites where the landfililing is almost completed, the retained water and pore water within landfill ground become high alkaline with pH of 11 or more. Therefore, the landfilling may be completed, while the pH is still high [2]. In order to use these sites, e.g., for factory or industrial land-use, after the landfilling is completed, the quality of the leachate generated during rainfall must satisfy the standard, e.g., pH of 9.0 or less. However, the pH of pore water in the landfilled ground remains high at most of the landfill sites, where the long-term treatments are required. This is because the alkaline components and organic components contained in the landfilled wastes become the pollution sources and those components will continue to release into the pore water in the landfill ground for a long time.

The countermeasures applied to purify the landfilled ground after the landfilling is complete such as “active washout using rainfall” and “forced drainage using a catchment underdrain” [3] were reported. Since the landfill sites were landfilled with various kind of wastes, the landfill ground may have different water permeability and air permeability. For that reason, although the purification can be promoted in the landfill ground with large grain size that makes it easy to flow through and ventilate, but it is not effective for the ground with small grain size due to its low permeability.

In such a case, in order to close the disposal sites, it is inevitable to choose uncertain or uneconomical countermeasures after the landfilling is completed.

As mentioned above, there is a need for a technology to quickly stabilize the leachate generated from the landfill ground, so that the disposal site can be closed early by implementing some countermeasures during the landfilling process, before the site is completed which makes it difficult to be purified. Therefore, in our research project, by focusing on the pH as the target of water quality, we assumed some prospective countermeasures that can be applied during the landfilling process. Those countermeasures are to maintain the buffering capacity of the retained water as much as possible before the
landfilling is completed.

Among those countermeasures, in this paper, we present only the countermeasure with the pumping up of the fresh seawater into the retained water in the landfill site to lower the pH and to enhance the buffering capacity of the retained water (See Figure 1).

In this paper, we attempted to control the pH of retained water in the coastal waste disposal site during the landfilling process, by conducting field surveys and two separated laboratory experiments.

## 2 Field surveys

### 2.1 Method

#### 2.1.1 Study site

The study site, namely, Port of Kinuura No. 3 Coastal Waste Disposal Site, is located in Taketoyo-cho, Chita-gun, Aichi prefecture, Japan (see Figure 2a). With the Waste Disposal Site, is located in Taketoyo-cho, Chita- total capacity of 496 Mm$^3$, the site is divided into 2 compartments for stable waste (8.4 ha) [4]. However, in this study, we focused on only the compartment for controlled-type waste which receive mainly fly ash. The landfill operation started to receive the waste in March 2011, and in March 2017, the volume of the landfilled waste was estimated to be about 25% of its total capacity.

#### 2.1.2 Survey locations and dates

In order to investigate the changes of water quality of the retained water in this landfill site, four times of surveys had been conducted in summer and winter seasons from 2015 [5] to 2017. Figure 2b shows the survey locations, one location (Point C) is at the central part where water depth is relatively deep and the other four locations are at the East, West, South, and North parts of the site, where the water depths are relatively shallow.

![Fig. 2. a) Study site location and aerial view and b) survey locations and water depth](image)

#### 2.1.3 Survey equipment

In the surveys, the vertical distribution of pH, water temperature, DO, and salinity were observed by using a Multi-Parameter Water Quality Sonde (YSI 6600 V 2-4). In addition, we collected water samples basically at a 2 m interval from the water surface to conduct the laboratory analysis for pH, bicarbonate ion (HCO$_3^-$), dissolved metals (e.g., calcium, magnesium, iron, and aluminium), salinity, and chloride ion concentration.

### 2.2 Results and discussion

There was no difference between the observation values and the analysis values of the same depth in all the survey locations for all investigation days. From these results, in all the survey locations are assumed to be uniform in horizontal plan of the whole landfill site. Therefore, in this paper, only the data at point C is presented. Figure 3 shows the observation results at point C. From the survey results of 2017/1/31, the graphs show the pH increased with progress of landfilling, which were almost 10 from the top surface to middle layers and over 10 in the bottom layer. As for water temperature, the thermocline (steep vertical temperature gradient) occurred at the depth of 5~6 m in summer, but due to mixing due to high temperature in winter, it was uniform except the bottom layer. Salinity increased with the progress of landfill, and in the bottom layer it exceeded 40‰, and salinity halocline (steep vertical salinity gradient) developed at about 12 m in depth. Due to the influence of this halocline, water temperature, DO of water mass of 2017/1/31 was formed in the state that is not vertically mixed.

Figure 4 shows the results of samples analysis of point C. HCO$_3^-$ decreased with the landfill progress, which was equivalent to less than a half of the value of general sea area (about 100 to 120 mg/L). Calcium as a whole was higher than general sea area (about 300 to 400 mg/L), and it was higher at the bottom layer. Magnesium was lower than the value of the general sea area (about 1000 to 1300 mg/L), and it was lower and lower at the bottom layer. The value also has decreased with the progress of landfill. According to the above results, it is considered that the chemical reactions as shown in the below chemical equations, modified from [1], occurred particularly in the vicinity of the bottom layer.

\[
\text{Ca(OH)}_2 \leftrightarrow \text{Ca}^{2+} + 2\text{OH}^- \quad (1)
\]

↑Supplied from ash  ↑Increase in seawater

\[
\text{HCO}_3^- \leftrightarrow \text{H}^+ + \text{CO}_3^{2-} \quad (2)
\]

↑Decrease in seawater  ↓Increase in seawater

\[
\text{OH}^- + \text{H}^+ \leftrightarrow \text{H}_2\text{O} \quad (3)
\]

↓Decrease in seawater  ↓Increase in seawater

\[
\text{Mg}^{2+} + \text{CO}_3^{2-} \leftrightarrow \text{MgCO}_3 \downarrow \quad (4)
\]

↓Decrease in seawater  ↑White deposit
3 Laboratory experiments

3.1 Background

Two different laboratory experiments were carried out, separately. In both experiments, we used exactly the same fly ash taken from the site and artificial seawater made with same initial pH of about 8.2. Immediately after receiving the sample, we carried the sieve analysis for particle size distribution of fly ash. However, as show in Figure 5, the particle size of fly ash used in in the first experiment ranged from 0.85 to 2 mm, while particle size of less than 0.25 mm in the second experiment. With the smaller particle size of fly ash, we expected to have higher pH in the second experiment. Vertical pipes which assumed to be representative of the real landfill site in 1 dimensional vertically. However, the pipe length used in the first experiment is shorter than that of the second experiment.

By using short pipes, the experiment could be easily carried out to investigate the pH changes corresponding to different cases of water injection. However, with short pipes, the pH decrease due to not only from the effect of

Fig. 3. Survey and analytical results of retained water quality (Point C)

Fig. 4. Analytical results of retained water quality (Point C)
the fresh seawater injected, but also from of CO₂ in atmosphere dissolved into alkaline seawater during experiment. Therefore, the second experiment was carried out with a longer pipe to investigate the depth of CO₂ influence and to clearly confirm the effect of fresh seawater injection.

The same pH meter (PH-230SDJ PE-21, Sato Corporation) has been used in both experiments. The pH meter can be used to continuously measure the pH at any depth by directly plunging into the seawater in the pipes.

### 3.2 Seawater injection experiment

#### 3.2.1 Method

As shown in Figure 6, we used three acrylic pipes with an internal diameter of 10 cm and height of 1 m. At first, 5.0 L (height 63.7 cm) of artificial sea water with the initial pH of about 8.2 was input into the acrylic pipes, and the vertical pH was measured at an interval of 5 cm from the overlying water surface. Next, fly ash of 2.0 kg -wet was added little by little for 16 minutes. After that, it was left to stand for 24 hours, next, the vertical distribution of pH of overlying water was measured. And then, we started the injection of new artificial seawater using a small vertical pipe attached into the inside of each acrylic pipe. As shown in Table 1, the addition of new seawater, equivalent to 1% (50 mL) and 4% (200 mL) of the used seawater quantity, was carried out continuously every 24 hours for 6 days. The injection height of each case was set as shown in Table 1.

#### Table 1. Experimental cases for seawater injection

<table>
<thead>
<tr>
<th>Seawater injection</th>
<th>Case 1</th>
<th>Case 2</th>
<th>Case 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ratio (%/day)</td>
<td>1</td>
<td>1</td>
<td>4</td>
</tr>
<tr>
<td>Volume (mL/day)</td>
<td>50</td>
<td>50</td>
<td>200</td>
</tr>
<tr>
<td>Injection height (cm)</td>
<td>70</td>
<td>40</td>
<td>40</td>
</tr>
</tbody>
</table>

**Fig. 6.** Experiment conditions

**3.2.2 Results and discussion**

The changes of vertical distribution of pH before and after injection of new artificial seawater as a measure to lower the pH of alkaline seawater are shown in Figure 7. From the results, in Case 1, where the water injection height was 70 cm from the bottom surface of the acrylic pipe (about 5 cm below the overlying water surface), the pH from the injection position to the water surface greatly decreased. Similarly, in Case 2 and Case 3 with injection height of 40 cm from the bottom surface of the acrylic pipe (10 cm on the deposit surface), the pH below the injection height tended to decrease. The tendency was more remarkable in Case 3 with greater water injection volume. The pH of overlying water just above the sediment surface was suppressed small in Case 3, where much water injection volume and low water inject height compared to Case 2 and Case1.

The surface of the ash deposit, after 24 hours of injection, was covered with white precipitate and increased with increasing seawater injection in all cases. From the above results, it is considered that, the nearer to the deposited ash layer, the higher density of overlying water. Accordingly, when the water with a relatively low density injected, it diffuses towards the water surface, while neutralized with only the shallow part of overlying water. Therefore, this result suggests that a deep injection of new seawater is effective to neutralize the retained water as a whole.

After the injection was completed, it was left to stand. On the 49th day after injection, the overlying water and the deposited ash pore water of Case 3 were sampled and analyzed. Pore water was obtained by using a solid-liquid centrifuge separator. The analytical results of artificial sea

**Fig. 5.** Particle size of fly ash used in the experiments
water before injection, overlying water, and pore water of Case 3 are shown in Table 2. Compared to the pH before being left to stand, the pH decreased as a result of atmospheric CO₂ dissolution from the overlying water surface, and the bottom layer was rising due to the influence of high alkaline pore water released from the ash deposit. The amount of HCO₃⁻ was decreased from the surface of overlying water to just above and within the ash deposit. Concentration of HCO₃⁻ in general sea water with the maximum pH of 8.25, is about 90 % at pH 9 and about 50 % at pH 10 (see Figure 8). From a certain proportion of the dissolved HCO₃⁻, the dissociated hydrogen ion H⁺ (and carbonate ion CO₃²⁻) is neutralized with OH⁻ derived from waste which can be considered to suppress the rising pH of seawater [1]. Compared to general sea water, the experimental value of HCO₃⁻ is small in the range of pH 8.2 to 9.5, which indicates that it was consumed by the neutralization reaction. At pH 10.5 it is considered that neutralization reaction does not occur because it is the same rate as general seawater. Therefore, the effect of injecting new seawater remarkably when the initial pH was 9.0 or more.

From the analytical results, the concentration of magnesium decreased downwards, and especially in the pore water. This is probably because the magnesium ion (Mg²⁺) in the overlying water or pore water react with CO₃²⁻ dissociated from HCO₃⁻ and form as precipitate of magnesium carbonate MgCO₃, resulting in a decrease in dissolved concentration [1]. It is thought that the increased in the sediment at the surface layer of the sediment is caused by further neutralization and bonding reactions as the pH had increased.

### 3.3 Effect of atmospheric CO₂ and seawater injection experiment

#### 3.3.1 Method

In order to make pH of the overlying water higher than the above experiment, smaller particle size of fly ash (under

---

**Table 2. Analytical results of artificial seawater, retained water, and pore water (Case 3)**

<table>
<thead>
<tr>
<th>Items</th>
<th>pH</th>
<th>HCO₃⁻ (mg/L)</th>
<th>Mg (mg/L)</th>
<th>Ca (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Artificial seawater (before input)</td>
<td>8.16</td>
<td>207.4</td>
<td>1300</td>
<td>640</td>
</tr>
<tr>
<td>Case 3</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Height of the overlying water from the pipe bottom (cm)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>85</td>
<td>8.29</td>
<td>116.1</td>
<td>1200</td>
<td>930</td>
</tr>
<tr>
<td>80</td>
<td>N.D.</td>
<td>N.D.</td>
<td>N.D.</td>
<td>N.D.</td>
</tr>
<tr>
<td>5</td>
<td>N.D.</td>
<td>N.D.</td>
<td>N.D.</td>
<td>N.D.</td>
</tr>
<tr>
<td>55</td>
<td>8.72</td>
<td>123.0</td>
<td>1100</td>
<td>1000</td>
</tr>
<tr>
<td>50</td>
<td>N.D.</td>
<td>N.D.</td>
<td>N.D.</td>
<td>N.D.</td>
</tr>
<tr>
<td>45</td>
<td>N.D.</td>
<td>N.D.</td>
<td>N.D.</td>
<td>N.D.</td>
</tr>
<tr>
<td>40</td>
<td>9.31</td>
<td>100.0</td>
<td>N.D.</td>
<td>2600</td>
</tr>
<tr>
<td>35</td>
<td>10.53</td>
<td>54.6</td>
<td>110</td>
<td></td>
</tr>
</tbody>
</table>
0.25 mm) was used. To clearly investigate the depth of influence of CO$_2$ dissolved into the overlying water and effect of the seawater addition, the use of a longer PVC pipe of 6 m in length and 75 mm in inner diameter was considered.

At first, as shown in Figure 9 the PVC pipe was firmly affixed to the wall of the experiment pit inside our research facility, where the environment is calm without strong wind. Then, about 24 L of artificial seawater with pH of 8.2 was injected into the PVC pipe. After that, 5 kg of fly ash with water content 12.36 % was input little by little. After 24, with an interval of 20 cm from the water surface, vertical distribution of pH of overlying water was measured.

As the injection method, a small PVC pipe was bended with 90 degree angle at the end so as not to allow the injected water drop below the exact depth, which was set to be about 2 m from the overlying water surface.

### 3.3.2 Results and Discussion

As a result, the average vertical distribution of pH of overlying water had been measured to be ranged from 11.0 to 11.5 for the first two months and a half, from the

![Fig. 8. Carbonate ion concentration of the general seawater and the experimental value](image)

![Fig. 9. Illustration of the second experimental condition](image)
beginning of the experiment. Based on the results during these period, the trend of vertical distribution of pH slightly decreased from the depth of 1 m downwards to the surface of overlying water. From this results, we could assumed that, under the effect of atmospheric aeration, CO$_2$ may dissolve into the overlying water during experiment until the depth of about 1 m from the overlying water surface.

After 3 months from the beginning of the experiment, the average pH of overlying water, measured from the surface to the injection depth of 2 m, was about 10.28. From then, we started to carry out the new artificial seawater injection. The first injection of about 220 mL of artificial seawater which was equivalent to about 1% of the total volume of the seawater initially used. After 24 hours, vertical distribution of pH was measured, and after that, immediately we carried out the second injection in the same way and the same amount of artificial seawater as the first injection. After 24 hours, the vertical distribution of pH was measured again. The results of vertical distribution of pH measured just before and after injection are shown in Figure 10.

We considered the average value of pH measured from the water surface to the injection depth of 2 with an interval of 20 cm. The average pH was about 10.28 just before the injection, had dropped to about 10.14 at 24 hours after the first injection, and then, dropped to 10.07 at 24 hours after the second injection. And, at 48 hours after the second injection the average pH very slightly decreased to just little below 10.07. Although the injection depth was at 2 m, however, the small effect of the seawater injection can be observed until the depth of 2.4 m. On the other hand, the pH distribution below the depth of 2.4 m was slightly increase. This may be due to the pore water with high alkalinity continued to release from the deposited ash layer as time pasted.

From these experimental results, especially, pH at 24 hours after the first injection, the effect of adding new artificial seawater in lowering pH of alkaline seawater was clearly confirmed.

4 Summary and concluding remarks

In Japan, coastal waste disposal sites are generally built with large scale and high construction cost. To use the sites after landfilling is completed for, e.g., industrial land-use, it requires much time and cost for stabilization of landfill ground (leachate treatment) to meet the standards. Therefore, our study aimed to control the pH of retained water in the coastal waste disposal sites during the landfilling process, by conducting field surveys and laboratory experiments.

Water quality surveys of the retained water were conducted for two consecutive years (in summer and winter) at the study coastal waste disposal site. In summer, a thermochemical layer and a halocline layer were confirmed, and in winter, the halocline of the bottom layer was confirmed. In such an environment that the halocline derived from the landfill waste is not eliminated throughout the full term, even if, for example, some of purification measures applied from the surface layer to the middle layer, because the purification effect may not reach the deep seawater layer with high density.

As the landfill progresses, the pH has risen to nearly 10, it is considered that the retained water in this study site reaches its buffering capacity limit [6] due to the decrease in HCO$_3^-$ in the retained water, the increase in dissolved calcium concentration in the bottom layer, etc.

In order to control of pH of retained water of the study site, we consider a low-cost method by simply pumping seawater from the adjacent sea into the site. The added fresh seawater is rich in HCO$_3^-$. Therefore, the H$^+$, dissociated from HCO$_3^-$, react with OH$^-$ eluted from landfill waste to result in lowering the pH. With this mechanism the buffering capacity of the retained water can be maintained. From the results of the laboratory experiments, this measure was confirmed effective for the alkaline seawater with pH of 9 or over. Therefore, the study suggests this method as one of the effective and low-cost methods applied during landfill process, so the disposal site can used promptly after the landfilling is completed.

![Fig. 10. Results of vertical distribution of pH before and after seawater injection](image-url)
The authors would like to thank ASEC (Aichi Seaside Environment Center) for allowing us to carry out research on this coastal waste disposal site. We are also grateful for its partial data provision for this paper.

References


[2] JESC (Japan Environment Sanitation Center), Examination survey toward the technical standard of the sea area final disposal sites for closure or abolished, pp. 18 (2005). (In Japanese, title is unofficially translated by authors)


