DEVELOPMENT OF NEW-TYPE LEACHATE TREATMENT TECHNOLOGY FROM LANDFILL SITES

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ABSTRACT
A new-type leachate treatment technology was demonstrated for a long time running performance. The technology includes not only the assured safety of treated water, but also the detoxification/stabilization and resource recovery/reuse of the by-products of treatment, such as sludge and concentrated/dried materials.

An experimental landfill was constructed. The new-type treatment facility attached with the coagulating membrane filtration unit, the desalination unit and the bipolar membrane electrodialysis unit to the conventional facility was introduced.

The experiments are divided into leachate treatment and resource recovery. The leachate treatment experiments resulted in chloride ion concentration lower than 200mg/L, regardless of the load variations due to seasonal variations of rainfall and the elapsed number of days since cells were filled with wastes. Also, the resource-recovery experiments showed that it was possible to recover approximately 1mol/L of acids and alkalis from desalination concentrates discharged from the leachate treatment process.

From these results we conclude that the new-type leachate treatment technology is helpful in constructing landfill sites acceptable to neighboring communities and thus contributing to environmental protection and the creation of a recycling society.

INTRODUCTION
Lately the problem of final waste disposal is a great national issue. Given a history of environmental pollution due to improper treatments and storage in the past, and the consequent mistrust of citizens, the development of “a final disposal site acceptable to communities” is awaited.
In order to realize the desirable leachate treatment, it is necessary to establish a "new-type leachate treatment technology." For such a new-type technology, the following points are considered to be important:

1. Concepts of securing the landfill site against ground contamination (reduction of the environmental risk)
2. Removal of inorganic salts from leachate and recovery of acid and alkali from desalination concentrates.

In consideration of the above, we prepared an experimental landfill and monitored the leachate. We also carried out leachate treatment experiments with a new-type leachate treatment facility, as well as acid and alkali recovery experiments from by-products (desalination concentrates).

**EXPERIMENTAL**

**Leachate treatment**

**Outline of the landfill:**

An experimental landfill (10m wide x 10m long x 2.5m deep) in the shape of a tapered cube was constructed. The landfill was filled to the height of 1.7m from the bottom in 5 days with a 6:4 (by volume) mixture of incineration plant ash and incombustible refuse. The surface was covered with approximately 0.2m fill soil. Photo 1 shows the landfill being filled with wastes.

**Treatment flow:**

A new-type leachate treatment facility was installed for experiments. Figure 1 shows a flow diagram of the system. The leachate from the landfill is collected in the leachate pit, controlled at a flow rate of 50L per day in the flow control tank, and passes thru an alkali coagulating sedimentation unit. In this unit sodium carbonate is added to remove calcium. The biological treatment, contact aeration unit decomposes, organic compounds followed by the coagulating ceramic membrane filtration unit where suspended solids are removed. The remaining trace amount of organic compounds and color are

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Photo 1. The experimental landfill being filled with wastes

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removed at activated carbon sorption unit, followed by the electrodialysis desalination unit. The by-product concentrate from the desalination unit was used for the resource recovery experiment described.

**Resource recovery**

**Outline of the bipolar membrane unit:**

As Fig. 2 depicts, bipolar membrane has a two-layer structure, an anion-exchange membrane and a cation-exchange one. A bipolar membrane is set so that the anion-exchange membrane faces anode and the cation-exchange one faces cathode. DC current is applied between the electrodes. The ions in water diffused into the membrane are exchanged to H⁺ and OH⁻ in the membrane. H⁺ drifts to the cathode and OH⁻ to the anode.

The bipolar membrane electrodialysis utilizes the aforementioned characteristics of this membrane. The system utilizes anion-exchange membranes and cation-exchange membranes similar to those in conventional electrodialysis systems for desalination, and bipolar membranes between them, forming a three-compartment structure. Figure 3 shows the mechanism of bipolar membrane electrodialysis.

![Fig 2. Mechanism of bipolar membrane](image)

**Fig 2. Mechanism of bipolar membrane**

![Fig 3. Schematic diagram of bipolar membrane electrodialysis](image)

**Fig 3. Schematic diagram of bipolar membrane electrodialysis**

![Fig 4. Flow diagram of experimental bipolar membrane electrodialysis system](image)

**Fig 4. Flow diagram of experimental bipolar membrane electrodialysis system**

![Photo 2. Appearance of the bipolar membrane unit](image)

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The experimental apparatus, a bipolar membrane electrodialysis unit for batch-wise operation was comprised of an electrodialysis tank (with five pairs of 0.02m² membrane), circulation systems for desalination concentrates (brine), acid solution, alkaline solution, etc. as shown in Fig. 4. Photo 2 shows the appearance of the unit.

**Set up of apparatus:**
The concentrates produced in the aforementioned experimental desalination process were used for this experiment. The chloride ion concentration in the concentrates was 31 000mg/L. The 4.0L brine was supplied to the brine tank of the bipolar membrane electrodialysis unit. Batch operation of the electrodialyzer was conducted until no acid/alkaline solution could be produced. In order to apply the DC current, 4.0L of 0.01 mol/L acid/alkali were added each to the acid/alkaline solution compartments in advance.

**RESULTS AND DISCUSSIONS**

**Leachate treatment:**

**Concentration of BOD and COD in leachate:**

Figure 5 illustrates the concentration change of BOD and COD in leachate during the experimental period. On the 26th day from the landfill set up the leachate was collected for the first time due to a rainfall. While the concentration of BOD and COD in leachate was estimated to be 250mg/L and 100mg/L, respectively, it marked a maximum of 2700mg/L and 1660mg/L, respectively on the 100th day. From the 160th day, the water quality started to decline gradually. On the 623rd day, when the experiment was finished, the value was as low as 2.2mg/L in BOD and 35mg/L in COD. During the period between 100th and the 300th days when leachate showed significantly higher concentration than estimated, the leachate was diluted with tap water to approximately 200mg/L in COD before feeding to the plant (hereinafter referred to as "plant raw water"). This indicates that the concentrated leachate flows out at an early stage, while uncondetrated one is discharged for a long time thereafter.

**Treated water quality:**

Tables 1 and 2 list the treated water quality of the new-type treatment facility. The plant raw water on the 273rd day with a BOD of 250mg/L was biologically treated to 8.0mg/L. BOD was then reduced to 0.2mg/L by coagulating membrane filtration.

**Table 1. Treated water quality at 273rd day**

<table>
<thead>
<tr>
<th>Analysis item</th>
<th>Plant raw water</th>
<th>Alkali coagulating filtration</th>
<th>Biological treatment</th>
<th>Coagulating membrane filtration</th>
<th>Desalination</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH (-)</td>
<td>7.5</td>
<td>7.3</td>
<td>8.9</td>
<td>7.0</td>
<td>7.0</td>
</tr>
<tr>
<td>BOD (mg/L)</td>
<td>250</td>
<td>180</td>
<td>8.0</td>
<td>0.2</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td>COD (mg/L)</td>
<td>110</td>
<td>90</td>
<td>50</td>
<td>24</td>
<td>8.6</td>
</tr>
<tr>
<td>T-N (mg/L)</td>
<td>27</td>
<td>16</td>
<td>14</td>
<td>14</td>
<td>1.7</td>
</tr>
<tr>
<td>SS (mg/L)</td>
<td>6.0</td>
<td>11</td>
<td>19</td>
<td>&lt;1.0</td>
<td>&lt;1.0</td>
</tr>
<tr>
<td>T-Ca (mg/L)</td>
<td>1500</td>
<td>5.6</td>
<td>4.6</td>
<td>5.7</td>
<td>1.9</td>
</tr>
<tr>
<td>Cl⁻ (mg/L)</td>
<td>4800</td>
<td>4300</td>
<td>4300</td>
<td>4400</td>
<td>180</td>
</tr>
</tbody>
</table>

**Table 2. Treated water quality at 560th day**

<table>
<thead>
<tr>
<th>Analysis item</th>
<th>Plant raw water</th>
<th>Alkali coagulating filtration</th>
<th>Biological treatment</th>
<th>Coagulating membrane filtration</th>
<th>Desalination</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH (-)</td>
<td>7.5</td>
<td>6.7</td>
<td>8.1</td>
<td>7.1</td>
<td>6.3</td>
</tr>
<tr>
<td>BOD (mg/L)</td>
<td>13</td>
<td>1.8</td>
<td>0.8</td>
<td>0.4</td>
<td>0.8</td>
</tr>
<tr>
<td>COD (mg/L)</td>
<td>52</td>
<td>35</td>
<td>31</td>
<td>16</td>
<td>2.8</td>
</tr>
<tr>
<td>T-N (mg/L)</td>
<td>11</td>
<td>7.8</td>
<td>5.4</td>
<td>3.8</td>
<td>2.1</td>
</tr>
<tr>
<td>SS (mg/L)</td>
<td>280</td>
<td>20</td>
<td>11</td>
<td>&lt;1.0</td>
<td>&lt;1.0</td>
</tr>
<tr>
<td>T-Ca (mg/L)</td>
<td>680</td>
<td>3.6</td>
<td>11</td>
<td>13</td>
<td>&lt;1.0</td>
</tr>
<tr>
<td>Cl⁻ (mg/L)</td>
<td>2200</td>
<td>2000</td>
<td>2000</td>
<td>2200</td>
<td>180</td>
</tr>
</tbody>
</table>
This shows that coagulating membrane filtration also has the function of reducing BOD. On the 560th day, the BOD was reduced from 13mg/L in raw water to 0.8mg/L at biological treatment. This is an example of the coagulating membrane filtration unit reducing also BOD by working simultaneously as an activated sludge treatment unit. The activated sludge treatment unit is realized by accommodating the outflow sludge from the biological treatment unit.

On the 273rd day the 110mg/L COD in plant raw water reduced to 24mg/L by coagulating membrane filtration, and to 8.6mg/L by desalination; 92% removal efficiency was achieved. On the 560th day, COD was 52mg/L in plant raw water, 16mg/L after coagulating membrane filtration, and 2.8mg/L after desalination; 95% removal efficiency was achieved. By using ferric chloride as the coagulating reagent under week acidic condition, COD removal ratio from biologically treated water was about 50%, which was almost satisfactory.

The T-N value was lower than 3mg/L at the end in desalinated water, regardless of the value in plant raw water. The T-N value did not decrease after the biological treatment because we incorporated only the nitrification unit relying on the subsequent desalination unit.

SS was bellow 1.0mg/L in the coagulating membrane filtration process both on the 273rd and the 560th days since it was filtered out by the ceramic membrane with ID 0.1um pore distributed.

A 99.5% of calcium was removed in the alkali coagulating sedimentation process, and 99.9% by the whole treatment system on both days.

The chloride ion concentration after the desalination process was 180mg/L on both days, lower than the standard value for tap water 200mg/L, regardless of the chloride ion concentration in the raw water. The results showed that the coagulating membrane filtration unit could reduce BOD, COD and SS while the desalination unit could reduce chloride ions and T-N; the new-type leachate treatment combining these units could reduce these species to safe levels for water environment, regardless of leachate load levels.

**Resource recovery**

**Recovery of acid and alkaline:**

Figure 6 shows the correlation between the bipolar membrane electrodialysis processing time and concentration of acid H+, alkaline OH and brine Cl-.

![Fig. 6. Correlation between bipolar membrane electrodialysis processing time and concentrations](image)

The concentration of by-produced acid and alkaline in the bipolar membrane electrodialysis unit was as low as 0.01mol/L at the beginning, whereas the concentration gradually increased to 0.88mol/L (chlorine 31 000mg/L). A 1.1mol/L of acid and a 1.2mol/L of alkaline were recovered from the concentrates. On the contrary, the chlorine in the treated water keep decreased. The treated water was desalinated to 0.005mol/L (chlorine 168mg/L) in the final stage. We could not recover the acid and the alkaline further since we reached the ending point. Figure 6 illustrates that the desalination takes place simultaneously as acid and alkaline recovery.
Moreover, the recovered acid and alkaline were concentrated enough to reuse at the leachate treatment facilities in landfill sites.

**Recovery of other resources:**
Several resources like nitrate and potassium ions present in the brine. Figure 7 outlines the results of the resource recovery, i.e., mass balance of chloride and sodium ions by bipolar membrane electrodialysis.

We found that 94.0% (118.7g) of chloride ions migrated from brine solution to acid solution in the experiment. Likewise a 89.6% (84.1g) of sodium ions migrated from brine to alkaline solution. Therefore, about 90% of the chloride/sodium ions migrated from brine to acid/alkaline solutions.

**SUMMARY**
From the results of the leachate-treatment/resource-recovery experiments, we obtained the following results;

1. The concentrated leachate outflowed at the beginning of the process and diluted with time.
2. The proposed leachate treatment facility could maintain chloride ion and SS concentration below 200mg/L and detection limit, respectively, regardless of the variation of leachate load.
3. With a bipolar membrane electrodialysis unit, 1.1 mol/L acid and 1.2 mol/L alkaline were recovered from desalination concentrates. The concentration of the acid and alkaline is well acceptable at leachate treatment facilities.
4. The desalination occurs at the same time as the acid/alkaline recovery. As a result, 168mg/L of chloride ion concentration in brine was achieved, which was lower than the standard value under the Quality Standard for Drinking Water (200mg/L).

The proposed technology can be realized in a system where brine is reduced and acid/alkaline are recovered as resources. This can facilitate the creation of a recycling society thru the conservation of the environment and the circulation of resources.

**REFERENCES**
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