High Gradient Magnetic Separation Combined With Electrocoagulation and Electrochemical Oxidation for the Treatment of Landfill Leachate

I. Ihara, K. Kanamura, E. Shimada, and T. Watanabe

***Abstract*—Landfill leachate was treated in a bench scale plant by high gradient magnetic separation combined with electrocoagulation using iron electrodes and electrochemical oxidation. The electrocoagulation with iron electrodes was used as a magnetic seeding method for phosphorus and organic compounds. Some organic compounds in landfill leachate was coagulated by iron(II) hydrates or hydroxides produced from iron electrodes. Phosphorus in landfill leachate was also suspended as iron phosphate. These magnetized solids were removed by high gradient magnetic separation using a superconducting magnet. The removal rate of total phosphate (T-P) was approximately 90% at a flow rate of 100 L/h. Residuals in the effluent of magnetic filter such as ammonium nitrogen ($NH_4 - N$) and chemical oxygen demands (COD) were destroyed by electrochemical oxidation using Ti/PbO_2 anode. The hypochlorite generated from oxidation of chloride in leachate might play a key oxidant in the effective removal of $NH_4 - N$. The experimental result showed that the pretreatment combined process of electrocoagulation using iron electrodes and high gradient magnetic separation might improve the charge efficiency in electrochemical oxidation. Several chemicals that were suspected of having environmental endocrine disrupting effects, was also decreased.**

***Index Terms*—Electrochemical oxidation, electrocoagulation, landfill leachate, magnetic separation.**

I. Introduction

LANDFILL leachate from municipal solid waste usually contains high concentrations of organic compounds and ammonium nitrogen, which are potentially major environmental hazards as surface and groundwater contamination [1]. Nitrogen or phosphorus loading is of concern to closed water bodies such as lake or bay because excess phosphorus or nitrogen can cause eutrophication. Approximately 190 organic compounds including chemicals suspected of having environmental endocrine disrupting effects were detected in landfill leachates in Japan [2]. Conventional biological wastewater treatment methods alone may not be effective in treating of landfill leachate as a result of refractory organic compounds. With increasingly requirements for pollution control from landfill leachate, the effective and economical treatment systems need to be developed [3]. In recent years, new physical or chemical wastewater treatments have gained increasing interest because they are relatively economical and give higher treatment efficiencies. Magnetic separations have potential that remove much magnetic particles from liquid, and its applications have been attempted for environmental remediation [4]. We constructed a bench scale wastewater treatment system consisted of electrocoagulation using iron electrodes, high gradient magnetic separation and electrochemical oxidation. The purpose of this study was to evaluate the feasibility of the treatment of nitrogen, phosphorus and refractory compounds including chemicals, which were suspected of having environmental endocrine disrupting effects in landfill leachate.

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II. Materials and Methods

A. Landfill Leachate

A landfill leachate sample was collected at waste disposal and land reclamation site inside the Tokyo metropolitan central breakwater in Tokyo bay. The landfill has been receiving industrial and domestic waste.

B. Design of the Wastewater Treatment System

The basic concept of the wastewater treatment system combined with electrocoagulation, high gradient magnetic separation and electrochemical oxidation is described in Fig. 1. The electrocoagulation using iron electrodes was used as magnetic seeding method for organics and phosphorus in landfill leachate. Magnetic substances generated by electrocoagulation were removed with magnetic separation using a superconducting magnet. The residuals in the effluent of magnetic filter such as ammonium nitrogen and refractory organics were destroyed by electrochemical oxidation using Ti/PbO_2 anode.

A schematic diagram of the wastewater treatment system used in this study was shown in Fig. 2. The leachate was pumped once into a plastic influent tank (400 L). The landfill leachate from the influent tank was continuously pumped to an electrocoagulation tank (40 L) at a constant flow rate of 100 L/h. Sheets of iron were used as the cathodes and anodes, and situated approximately 30 mm apart from each other and dipped

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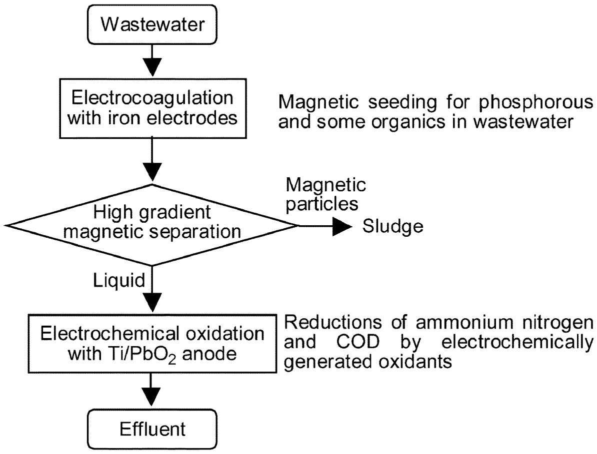

Fig. 1. Basic concept of wastewater treatment system for landfill leachate.

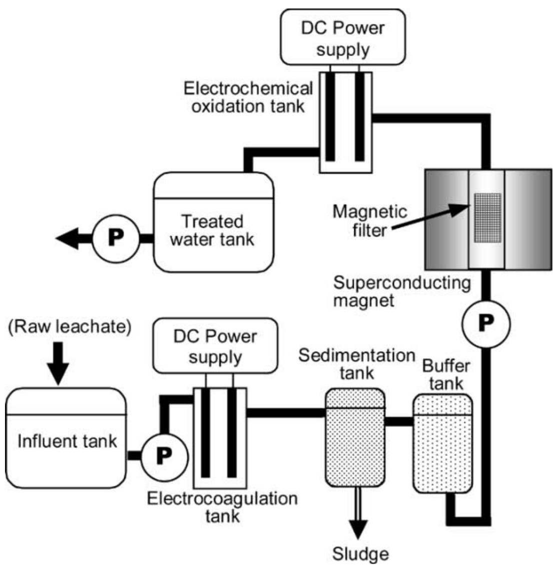

Fig. 2. Schematic diagram of the wastewater treatment system for landfill leachate.

Fig. 3. View of the wastewater treatment system with a superconductor magnet.

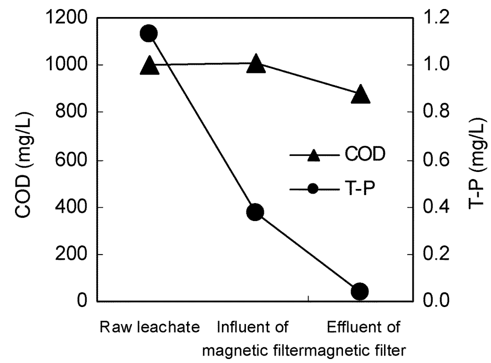

Fig. 4. Reduction of COD and total phosphate (T-P) for the electrocoagulation and magnetic separation.

into the leachate. The electrolytic current was 35 A supplied by a DC power supply (Takasago, EX-1500L2). From the electrocoagulation tank, it was flowed out to a sedimentation tank (40 L), and then transported for subsequent purification to a buffer tank (40 L). Part of suspended solids was removed in the sedimentation tank, where they precipitated to the bottom as sludge. The solids from the bottom of sedimentation tank were handled as sludge. To maintain the solids content in the optimal range, sludge wastage was carried out periodically. The supernatant from the buffer tank was then pumped through a superconducting magnet (Japan Superconductor Technology, JMTD-10T100SS) to remove suspended solids. The magnetic separation was performed with high gradient magnetic filters in a high gradient magnetic field. The magnetic filter consisted of tightly rolled wire meshes (Type 430 stainless steel) and set along a fluid flow. An acrylic cylindrical chamber with magnetic filters was fixed inside the bore. The external magnetic field at 10 T supplied by a superconducting magnet was the same direction with liquid flow. The effluent of magnetic filter was flowed into the electrochemical oxidation tank (30 L). Meshes of Ti/PbO2 were used as anodes and Sheets of Ti was used as cathodes in the electrochemical oxidation. The current density was kept constant by using a DC power supply (Takasago, EX-1500L2). The experiment of electrochemical oxidation was operated in batch mode.

III. Results and Discussion

A. Electrocoagulation and Magnetic Separation

Fig. 4 shows the reduction of COD and total phosphate (T-P) from landfill leachate by electrocoagulation using iron electrode and magnetic separation at a flow rate of 100 L/h. The concentration of COD in the raw leachate was 1000 mg/L, and reduced to 880 mg/L in the effluent of magnetic filter. The mechanisms in the electrocoagulation process include coagulation and adsorption, absorption, precipitation and floatation [5]. Some organic compounds in landfill leachate adsorbed on iron(II) hydrates or hydroxides generated by electrocoagulation using iron electrodes. This result indicated that the COD was reduced by removing these hydrates or hydroxides in the magnetic separation. On the other hand, the removal efficiency of 96.6% was obtained after magnetic separation. Because phosphorus in the landfill leachate was mainly suspended as iron phosphate by electrocoagulation. The result shows that some iron phosphate was precipitated in the sedimentation tank and nonsedimentable iron phosphate was removed by magnetic separation.

B. Electrochemical Oxidation Process

The experimental result of the electrochemical oxidation of the magnetic filter was shown in Fig. 5. The value of current

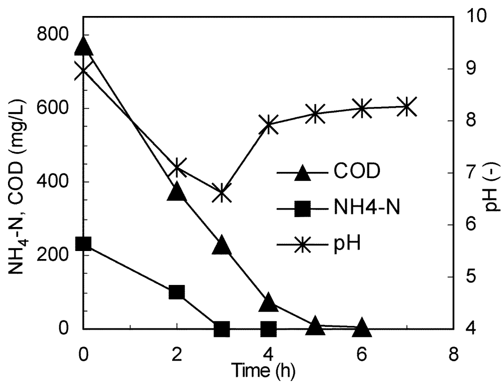

Fig. 5. The result of electrochemical oxidation of the effluent of magnetic filter.

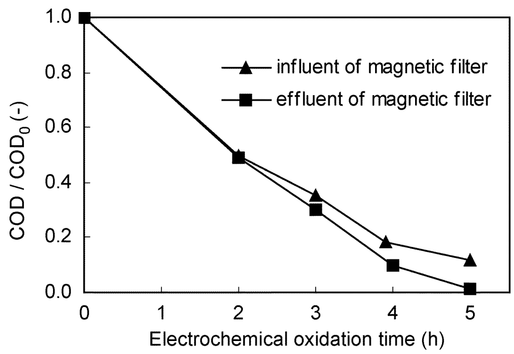

Fig. 6. Effect of magnetic filter for normalized COD during the electrochemical oxidation.

was 80 A in the experiment. The concentration of COD was decreased to 10 mg/L for 5 hours. In general, organic compounds in wastewater can be destroyed electrochemically by conducting a direct anodic oxidation or an indirect oxidation process [6]. The direct oxidation took place on the surface of the anode and the destruction of organic compounds might be attributed to the strong oxidation ability of the hydroxyl radical. The indirect oxidation was worked by hypochlorite, which was generated electrochemically from chlorides in wastewater. The landfill leachate was involved much chloride ion. Fig. 5 showed that the concentration of $NH_4 - N$ was decreased dramatically due to the indirect oxidation. The reduction mechanism of $NH_4 - N$ was similar to the "Selleck-Saunier Breakpoint Chemistry" [7]. Both reductions of COD and $NH_4 - N$ tended to relate the electrical charge during the experiment. The effect of magnetic separation in the electrochemical oxidation was shown in Fig. 6. The rate of COD reduction of magnetic filter effluent was higher than that of the filter influent. It was indicated that the removing of suspended solids by magnetic separation allow to improve the efficiency of COD reduction in the electrochemical oxidation.

C. Removals of Chemicals Suspected of Having Environmental Endocrine Disruptors

Bisphenol A, aniline and di-(2-ethylhexyl) phthalate were chemicals suspected of having environmental endocrine disrupting effects. The chemicals were detected in the landfill leachate used in this study. Fig. 7 showed the experimental results for the removals of the chemicals. In this experiment, the electrochemical oxidation was operated for 9 hours under

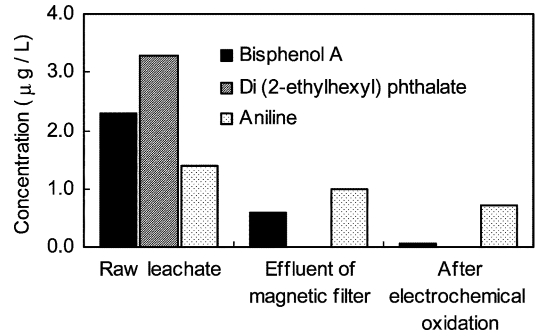

Fig. 7. Degradation of endocrine disturbing chemicals by electrocoagulation, magnetic separation and electrochemical oxidation.

50 A. After the electrocoagulation and magnetic separation, Bisphenol A and anile were removed and di-(2-ethylhexyl) phthalate was not detected. Bisphenol A and anile were more decreased during the electrochemical oxidation.

IV. Conclusion

The landfill leachate was treated by electrocoagulation, magnetic separation and electrochemical oxidation. The combination of electrocoagulation and magnetic separation could be performed effectively for removing of phosphorus in landfill leachate. The result suggested that the electrocoagulation using iron electrodes was suitable for magnetic seeding method of phosphorus. High treatment removals of $NH_4 - N$ and COD were obtained in the electrochemical oxidation using Ti/PbO_2 anode. The reduction of $NH_4 - N$ was mainly due to indirect oxidation by hypochlorite generated from oxidation of chloride in the leachate. The result showed that the combined treatment for landfill leachate could be performed without addition of chemicals.

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