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Combined Adsorption and Chemical Precipitation Process for Pretreatment or Post-Treatment of Landfill Leachate

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Abstract−Performances of combined adsorption and chemical precipitation were evaluated as one of the options for pretreatment or post-treatment of a municipal solid waste (MSW) landfill leachate and leachate from an industrial solid waste landfill. The COD and color removals of the leachate from a MSW landfill were 35% and 33% at an alum dose of 300 mg/L with preceding PAC (powdered activated carbon) dose of 200 mg/L, respectively. For MSW leachate, the combined adsorption and coagulation process showed 2.3 times higher COD removal at PAC dose of 200 mg/L and alum dose of 500 mg/L than the unit process of adsorption with poor settleability. The COD removal was accomplished mainly by adsorption, while coagulation was a key mechanism of color removal. The COD and color of the biologically treated leachate from an industrial solid waste landfill were removed up to 32% and 68%, respectively, at addition of 490 mgAlum/L and 1,000 mgPAC/L in adsorption-coagulation process with pH control. Combined adsorption and coagulation process with pH control showed better COD and color removal than the process without pH control. The color removal was influenced greatly by pH control, while COD removal was not. No difference in removal efficiency was observed between adsorption-coagulation and coagulation-adsorption. Maximum net increases in the COD and color removals by the adsorption-coagulation process were 40% and 46%, respectively, compared with the removals by sole chemical precipitation. The Freundlich isotherm exclusively described the adsorption of leachate components on the PAC. Thus, a combined adsorption and coagulation process was considered to be effective for pre-treatment or post-treatment of landfill leachate, and has distinct features of simple, flexible, stable and reliable operation against fluctuation of leachate quality and flowrate.

Key words: Adsorption, Chemical Precipitation, Leachate, Pre or Post-treatment

INTRODUCTION

Landfill leachate contains a considerable amount of non-biodegradable organics, heavy metals, and unexpected hazardous materials such as ethylene, tetrachloroethylene and has intense color. Moreover, flowrate, BOD_5, CODcr and Color of leachate also fluctuate seriously. It is, therefore, strongly suggested that provisions against such a feature of leachate should be made to protect the main process against any potential hazards and shock loading from the leachate, and satisfy the effluent guideline. Pretreatment of leachate is usually required to prevent deterioration by an unexpected high COD loading as well as hazardous substances from solid waste landfill. Biologically treated leachate containing high concentration of non-biodegradable organics and intense color may also require post-treatment.

The physico-chemical processes have been used both for raw leachate with low content of biodegradable organics and for biologically treated leachate, which include adsorption, chemical precipitation, chemical oxidation and reverse osmosis [Kapetanios et al., 1995; Gau and Chang, 1996]. Activated carbon adsorption has been proven to be an effective process for leachate from landfill at the condition of methanogenic phase [Ehrig, 1989]. Research on the adsorption process treating a municipal solid waste (MSW) landfill leachate demonstrated a COD removal efficiency as high as 50% [Cossu et al., 1992]. Granular activated carbon (GAC) and powdered activated carbon (PAC) can be both used to remove organic components from wastewater and water. GAC has inherent advantages of applying activated carbon in fixed beds, including lower carbon usage rate for much application, and ease of spent carbon regeneration. However, PAC has the advantages of being a cheaper material and requiring minimal capital expenditure for feeding and contacting equipment, and it can be also applied only when needed [Najm et al., 1991]. Thus, the PAC adsorption process could be applied to treat leachate, although poor solid-liquid separation is a disadvantage of the process. Chemical precipitation was known to be an unsuitable treatment method for high strength leachate, while the process applied to methanogenic phase leachate reduced COD, AOX, suspended solids and color within a limited extent [Ehrig, 1989].

The objective of this study was to evaluate the feasibility of combined PAC adsorption and chemical precipitation with advantages of both processes as one of the options for pre-treatment of leachate from a municipal solid waste landfill and post-treatment of biologically treated leachate from an industrial solid waste landfill.

MATERIALS AND METHODS

Raw leachate was taken from an MSW landfill site (Nanjido)

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Table 1. Characteristics of the leachate [(range), average±standard deviation]

Parameters	Municipal leachate	Industrial leachate
pH	(7.5-8.6) 8.0±0.4	(7.6-9.3) 8.1±0.6
BOD_5	(60-340) 230±23	(2200-4200) 3220±650
COD_{Cr}	(1080-4750) 2400±200	(5100-8300) 6900±1120
Suspended solids	(11-67) 33±4.0	(280-1770) 630±570
Total Kjeldahl nitrogen	(1480-3350) 1780±70	(155-274) 215±45
Total phosphorus	(2.9-15.7) 7.3±1.6	(1.1-5.0) 2.2±1.4
Alkalinity(as $CaCO_3$)	(3790-8840) 6740±800	(1300-2300) 1690±360
Chloride	(1680-4120) 2430±360	(1970-6800) 5200±1760
Conductivity	-	(5200-17800)13800±4430
Color (ADMI value)	(1450-1480) 1470±30	(1980-2420) 2280±60
Oil & Grease	(0.02-0.82) 0.23±0.04	-
Cadmium	(0.01-0.05) 0.02±0.004	(0.33-1.18) 0.56±0.34
Chromium	(0.57-1.6) 0.95±0.06	(1.73-2.66) 2.04±0.44
Copper	(0.02-0.25) 0.09±0.004	(0.33-1.51) 0.94±0.42
Iron	(0.56-12.2) 4.4±0.5	(1.51-34.2) 18.2±12.3
Lead	(0.003-0.19) 0.0035±0.0	(4.37-11.0) 8.35±2.83
Zinc	(0.01-0.2) 0.13±0.03	(1.05-5.28) 3.48±1.66

All parameters except pH, conductivity and color were listed on the basis of mg/L.

closed in order to investigate the feasibility of an on-site pre-treatment prior to combined leachate treatment with sewage at a nearby municipal wastewater treatment plant, which required a reliable pre-treatment option before receiving the leachate. The raw leachate had a BOD_5/COD_{Cr} ratio of 0.096 relatively lower than other MSW leachate since this leachate was generated from old landfill site. Biologically treated leachate from an active industrial solid waste landfill was also used to evaluate effect of post-treatment. Average BOD_5/COD_{Cr} ratio of the biologically treated leachate was 0.32. The COD_{Cr} of raw leachate and biologically treated leachate averaged 2,400 mg/L and 1,100 mg/L, respectively. Table 1 illustrates typical characteristics of the leachate used in this study.

A PAC adsorption experiment with raw leachate from an MSW landfill was performed to select applicable PAC concentrations prior to an experiment on combined adsorption and chemical precipitation (coagulation). Under chosen PAC concentrations, a combined adsorption and coagulation experiment was carried out at various concentrations of coagulant. PAC smaller than 100 mesh and aluminum sulfate (alum) was used as adsorbent and coagulant, respectively. Adsorption and coagulation process proceeded as a series of contact mixing with PAC for 20 minutes, rapid mixing (140 rpm) for 5 minutes after addition of alum, slow mixing (30 rpm) for 15 minutes, and 2-hour settling at a temperature of 25 °C. All experiments were performed three times at the same conditions, and research results obtained from these experiments are presented in this article based on the average value.

Based on the operating conditions of an existing field chemical precipitation process for biologically treated industrial leachate, the effect of PAC addition to the chemical precipitation process was investigated to improve the existing field process at PAC dosage ranging from 100 to 2,000 mg/L. In order to investigate differences in COD_{Cr} and color removal with points of PAC addition, the performance of the adsorption-coagulation process and the coagulation-adsorption process was evaluated with and without pH control. The one was that PAC adsorption took precedence over coagulation, while the other was that coagulation was performed before PAC adsorption. For a combined adsorption and coagulation process with pH control, adsorption was performed at pH 8 with a 10-minute rapid mixing (140 rpm), and coagulation was accomplished at pH 4 with a 3-minute rapid mixing (140 rpm) followed by a 10-minute slow mixing (50 rpm), and 2-hour settling at pH 9. Sulfuric acid and sodium hydroxide were used for pH adjustment. Supernatant after settling was analyzed to evaluate effluent quality. All analyses in this study were conducted as per procedures in the APHA Standard Methods [APHA, 1995].

RESULTS AND DISCUSSION

1. Adsorption and Chemical Precipitation for Pretreatment

Combined adsorption and coagulation was evaluated as one of the options for pretreatment of a MSW landfill leachate. PAC adsorption and subsequent settling of PAC achieved removal of raw leachate COD. The COD_{Cr} removal had a tendency to increase from 20% to 45% at PAC dosage ranging from 100 to 1,000 mg/L; however, consistent COD removal at any PAC dosage could not be accomplished even with one-day settling of PAC after adsorption because organic adsorbates on carbon particles remaining in the supernatant could cause high COD exertion. Stable COD removals were achieved with improved settleability of PAC by chemical precipitation using alum. Two hours were enough for settling of flocculated PAC. Contact time for adsorption between 5 to 90 minutes did not affect the adsorption.

Fig. 1 illustrates the COD and color removal by combination of adsorption and coagulation process at fixed PAC dosages of 100 and 200 mg/L chosen based on the COD removal as well as in the economic aspect. The COD removal did not change significantly at an alum dosage over 100 mg/L with a preceding adsorp-

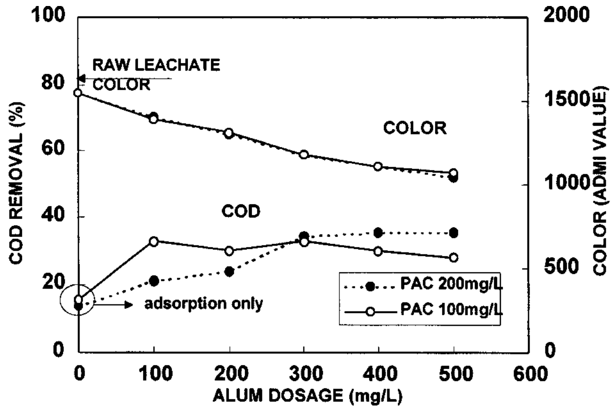

Fig. 1. The COD and color removal by combined adsorption and coagulation of an MSW landfill leachate.

tion by PAC dosage of 100 mg/L, while gradual increase in COD removal was observed as alum dosage increased with a preceding PAC dosage of 200 mg/L. Lower COD removal at higher preceding PAC dosage at an alum dosage below 300 mg/L was due to insufficient alum dosage to precipitate larger amount of carbon particles. Settleability of the chemical sludge increased with increasing alum dosage. Satisfactory zone settling occurred at an alum dosage higher than 300 mg/L regardless of PAC dosage.

Fig. 1 demonstrates that the COD removal was accomplished mainly by PAC adsorption rather than alum coagulation at the range of PAC dosages investigated, although flocculation to improve settleability of PAC resulted in an improved COD removal at higher PAC dosage. Adsorptive capacity of PAC was 5.5 to 6.5 mgCOD per mgPAC at a PAC dosage of 100 mg/L. Combined adsorption and coagulation process showed 2.3 times higher COD removal at PAC dose of 200 mg/L and alum dose of 500 mg/L than the unit process of adsorption with poor settleability. Identical linear color removal of a unit ADMI value per mg alum added was obtained with increasing alum dosage regardless of PAC dosage. As alum dosage increased at any PAC dosage, effluent luminance increased, and purity decreased, while the predominant yellow hue did not change. The color was removed principally by coagulation.

The removal of COD and color was 35% and 33% at an alum dosage of 300 mg/L with a preceding PAC dosage of 200 mg/L, respectively. Such a removal and its stability were considered to be effective for an on-site pre-treatment prior to combined leachate treatment with sewage at a field municipal wastewater treatment plant of interest. The activated sludge process was also operated to compare with the adsorption-coagulation process. The activated sludge process almost failed after 4 months operation, since activated sludge could not be acclimated to the raw leachate from the MSW landfill.

2. Adsorption and Chemical Precipitation for Post-Treatment

Combined adsorption and coagulation was examined to improve the performance of an existing field chemical precipitation process treating a biologically treated leachate from an industrial landfill. Fig. 2 shows the COD and color removal by combined adsorption and coagulation process at a fixed alum dosage of 490 mg/L applied in the field plant without pH control. The COD removal linearly increased from 13% to 51%, and the color removal did from 22% to 65% at PAC dosages ranging from 100 mg/L to 2,000 mg/L by adsorption-coagulation process without pH control. The COD and color removal mainly depended on adsorption rather than coagulation. Adsorptive capacity of PAC was 1.6 mgCOD per mgPAC, which was lower than that of raw MSW leachate, at a PAC dosage of 100 mg/L. Maximum net increase in COD and color removal of the biologically treated industrial leachate by adsorption-coagulation process were, respectively, 45% and 46% compared with the unit process of coagulation. Effluent from adsorption-coagulation process exhibited an excellent quality of color. No noticeable difference in removal efficiency was observed between the adsorption-coagulation process and coagulation-adsorption process, as shown in Fig. 2.

The COD and color removal by combined adsorption and coagulation process with pH control are presented in Fig. 3. The COD removal linearly increased from 21% to 51%, and the color removal did from 57% to 77% at PAC dose ranging from 100 mg/L to 2,000 mg/L with an alum dosage of 490 mg/L by adsorption-coagulation process. Adsorptive capacity of PAC was 2.4 mgCOD per mg PAC at a PAC dosage of 100 mg/L by adsorption-coagulation process. Combined adsorption and coagulation process with pH control showed better COD and color removal than the process without pH control. The pH control dur-

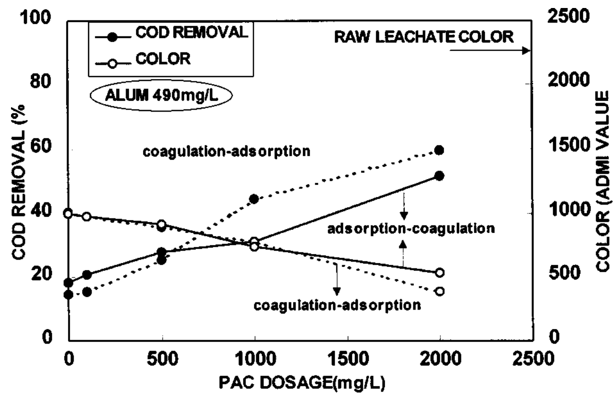

Fig. 2. The COD and color removal by combined adsorption and coagulation of a biologically treated industrial landfill leachate without pH control.

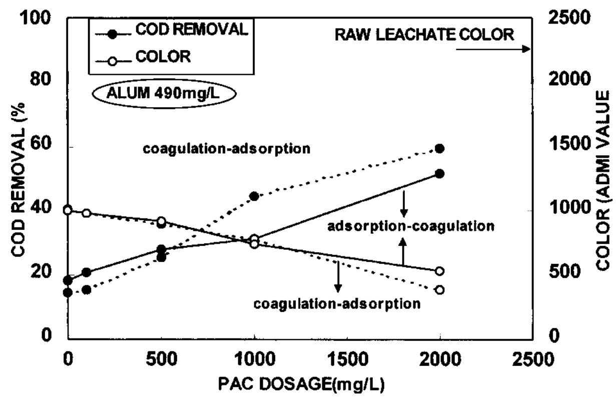

Fig. 3. The COD and color removal by combined adsorption and coagulation of a biologically treated industrial landfill leachate with pH control.

ing the combined adsorption and coagulation process did not significantly affect COD removal. Color removal was, however, significantly improved by pH control with acidification during coagulation. The point of PAC addition did not affect the COD and color removal in combined adsorption and coagulation with pH control, and this was clearly supported by Kim et al. [1999].

Thus, the combined adsorption and chemical precipitation process was believed to be reliable to enhance and stabilize the performance of the existing field chemical precipitation process treating biologically treated leachate, and to satisfy effluent guidelines.

3. Adsorption Isotherms

The adsorption phenomena between the liquid (leachate) and the solid phase (PAC surface) were analyzed by using the Langmuir, Freundlich and BET isotherms. Comparing plots of the amount adsorbed (mgCOD/gPAC) as a function of the equilibrium solution concentration for the models, the Freundlich isotherm exclusively described the adsorption of leachate components on the PAC. The Freundlich isotherm constants for raw leachate from an MSW landfill and biologically treated leachate from an industrial solid waste landfill are presented in Table 2.

The parameter K_f represents a specific capacity, an indicator of sorption capacity at a specific solution phase concentration. Although the specific capacities in Table 2 are lower than those of most pure organic adsorbates, the specific capacities are similar to those of other MSW leachates in the order of magnitude from 10^{-4} to 10^{-7} [Cossu et al., 1992]. The specific capacity of raw leachate was higher than that of biologically treated leachate. This indicated that the quantity of raw leachate components adsorbed was larger than that of biologically treated leachate. The 1/n value (adsorption intensity), as determined from the slope of the log-log plot of adsorption data, denotes a joint measure of the cumulative magnitude and diversity of energies associated with a particular adsorption reaction. Efficient adsorbents have the 1/n values ranging from 0.3 to 0.95 depending on the chemical nature of adsorbate. However, the adsorption intensities of the leachates investigated are higher than 1, which indicates concave isotherms, and those of other MSW leachates are also approximately ranging from 1 to 4 [Cossu et al., 1992]. Since raw leachate had lower adsorption intensity than biologically treated leachate, the adsorbent for biologically treated leachate could lose its capacity more rapidly than that for raw leachate as influent concentration is reduced to effluent concentration. When equilibrium concentrations increase, the effect of adsorption intensity becomes greater than that of specific capacity as demonstrated in Fig. 4.

Table 2. Freundlich isotherm constants

Adsorbate	K_f	1/n
Raw leachate from a MSW landfill	1.35×10^{-3}	1.92
Biologically treated leachate from an industrial landfill	3.15×10^{-5}	2.49

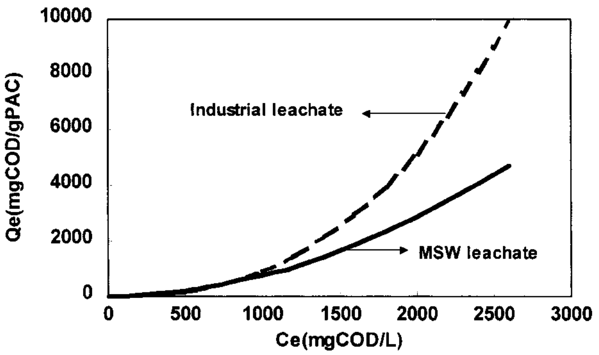

Fig. 4. The Freundlich isotherms for the leachates investigated.

CONCLUSIONS

The feasibility of the combined PAC adsorption-coagulation process was evaluated as one of the options for pretreatment of raw leachate from a municipal solid waste (MSW) landfill, and post-treatment of biologically treated leachate from an industrial solid waste landfill.

The combined adsorption and coagulation process for MSW leachate showed higher COD removal than the unit process of adsorption with poor settleability. The COD removal was accomplished mainly by adsorption, and color was removed principally by coagulation. For biologically treated industrial leachate, maximum net increases in COD and color removals by the adsorption-coagulation process were 40% and 46%, respectively, compared with the removal by sole chemical precipitation. The color removal was influenced greatly by pH control, while COD removal was not. No noticeable difference in removal efficiency was observed between the adsorption-coagulation process and coagulation-adsorption process.

The Freundlich isotherm exclusively described the adsorption of leachate components on the PAC. The isotherm showed that the adsorbent for biologically treated leachate loses its capacity more rapidly than that for raw leachate.

Combined adsorption and chemical precipitation process was considered to be an effective on-site pretreatment option for protecting a municipal wastewater treatment plant receiving MSW leachate against any potential organics shock loading. This process was also one of the efficacious options for post-treatment after biological treatment of leachate from an industrial solid waste landfill. The process could have the capability of coping with an extremely high shock loading by higher removals at higher chemical dosage. The adsorption and chemical precipitation has distinct features of simple, flexible, stable, and reliable operation against fluctuation of leachate quality and flowrate, and could be used as pre- and post-treatment, or upgrading option.

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