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ADVANCED OXIDATION TREATMENT OF THE LEACHATE COLLECTED FROM WASTE DISPOSAL LANDFILL SITE IN HANOI, VIETNAM – APPLICATION AND ITS EFFECTS

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ABSTRACT

The quality of leachate collected from waste landfills which have received the industrial and municipal waste in Hanoi were analyzed in order to obtain the information of the current situation of the sites. The leachate collected from Tay Mo landfill showed the high concentration of lead, iron and mercury and that from Nam Son landfill showed the high concentration of lead, iron, mercury and arsenic. It was also recognized that the high strength of COD and color of leachate would be primary problems to be resolved. The application of several advanced oxidation processes (AOPs) to Tay Mo landfill leachate revealed that Fenton reaction and photo-Fenton reaction under pH adjustment effectively removed COD and color from leachate and degraded the humic substance in the leachate. Mutagenicity of leachate was increased by UV-TiO_2-H_2O_2, Fenton (pH 3) and photo-Fenton (pH 3) treatment. It represents that degradation of organic matters in leachate resulted in generation of toxic intermediates. Further investigation for operating condition of AOPs treatment should be necessary to minimize the environmental impact of waste landfill leachate.

KEYWORDS

Advanced oxidation; COD removal; decolorization; landfill leachate; mutagenicity; Vietnam

INTRODUCTION

Vietnamese economy began to be developed rapidly in 1990's, and intensively industrial growth has caused serious environmental pollution especially in urbanized area such as Hanoi and Ho Chi Minh City. Increasing the generation of industrial and municipal wastes is especially going to be one of the greatest public concern. Waste landfill leachate has been reported as a pollution source of water environment in all over the world (Christensen and Christensen 2000). Especially in the case of Hanoi, environmental deterioration by landfill leachate is a quite serious problem because it has been recognized that groundwater has been greatly contaminated. It is, especially, life threaten problem in Hanoi because groundwater has been used for drinking water resources in most of residential area. Very few field survey of environmental pollution by leachate in Vietnam have been made and intentional study should be necessary to reveal the situation of the pollution. In this study, we described the current situation of leachate collected from waste landfill sites in Hanoi. Then advanced oxidation processes (AOPs), TiO_2-UV, TiO_2-UV-H_2O_2, Fenton reaction (w/ and w/o pH adjustment to 3) and photo-Fenton reaction (w/ and w/o pH adjustment to 3) were implemented for leachate treatment in order to remove COD and color. Numerous investigations have proven that organic compounds can be oxidized by AOPs which produce highly reactive OH□. The processes have been intensively investigated for the treatment of industrial wastewater and waste landfill leachate, which shows high COD/BOD ratio and heavy color. For successful leachate treatment by AOPs, the efficiency and operating condition of the AOPs were evaluated and discussed.

MATERIALS AND METHODS

Waste landfill site and sample collection

Tay Mo waste landfill site (5 ha) is located 10 km south-west from the central Hanoi area. The site had been managed by Urban Environment Company, the representative for solid waste management for the urban area of Hanoi, since 1997. This site has been fulled and closed in the early 2000. Nam Son waste landfill site (13 ha) was constructed by Hanoi People's Committee. It was located 50 km north-east from central Hanoi area. This site started to receive waste since 1999. Leachate and surface water around the both landfill sites were collected and provided for water quality analyses and various oxidation processes.

Experimental apparatus and operation of advanced oxidation process

AOPs were performed in photoreactor using a glass cylinder of 2-L volume. This reactor was operated in recirculation mode using a tubing pump. The 250 W UV lamp was served as a UV source and irradiated from inner surface of the annulus. Illumenite ($FeTiO_3$) which adsorbed and sintered onto activated carbon was served as a photocatalyst. As the advanced oxidation processes, TiO_2-UV system, TiO_2-UV-H_2O_2 system, Fenton reaction system (w/ and w/o pH adjustment to 3) and photo-Fenton reaction system (w/ and w/o pH adjustment to 3) were adapted for treatment of raw leachate. In the UV-TiO_2 process, 1 mg /l of catalyst was put to the annulus which was covered by mesh and influent was inlet from inside of annulus. In UV-TiO_2-H_2O_2 process, 2.5 ml of 50% hydrogen peroxide was added and operated at same condition as UV-TiO_2 system. In Fenton reaction process, Mohr's salt ($(NH_4)_2Fe(SO_4)_2 \cdot 6H_2O$) was added at the concentration of 2.5×10^{-3} mol/l as Fe (II) source. After the addition of 2.5 ml of 50 % hydrogen peroxide to the leachate, operation was started. Photo-Fenton process was operated under the same condition as Fenton process except UV irradiation. Adjustment of pH was carried out by addition of sulfuric acid. All effluents were left for 30 minutes for settlement and analyzed.

Coagulation

Raw leachate and effluents after the TiO_2-UV, TiO_2-UV-H_2O_2, Fenton reaction and photo-Fenton reaction under no pH adjustment were served for the coagulation. Mohr's salt was added at the concentration of 2.5×10^{-3} mol/l as a coagulant. After 30 minutes settlement, supernatant was used for water quality analysis.

Analytical procedure

Water quality analyses were conducted in accordance with the Japanese Industrial Standards K0102 (1995). Chemical oxygen demand (COD) was measured as dichromate oxygen demand (COD (Cr)). The concentration of heavy metals was analyzed by atomic adsorption spectrophotometer (Shimadzu AA6800). Distribution of molecular weight of humic substances in the leachate was analyzed by gel permeation chromatography (GPC) with Sephadex G-50 (Pharmacia LKB., Uppsala, Sweden) with an eluent of 50 mM sodium phosphate buffer (pH 7.0) at a flow rate of 1.5 ml/min.

Bioassay

Mutagenicity of each sample was evaluated by *umu* test (Oda *et al.*, 1987) using *Salmonella typhimurium* TA1535 harboring recombinant plasmid pSK1002 containing a fused gene *umuC'-lacZ'*. The leachate was confirmed as mutagenicity positive when the sample showed dose-respondent and two-fold increase of β-galactosidase activity compared to the background level. Estrogenic activity was evaluated by yeast two-hybrid method using yeast screen Y190 harboring GAL activation domain fused to the coactivator TIF1 and GAL4 DNA binding domain fused to ligand-binding domain of estrogen receptor (Nishikawa *et al.*, 1999).

Table 1 Quality of leachate and surface water taken from Tay Mo and Nam Son waste landfill area. Vietnamese standards for surface water and industrial water were also shown.

		Ni	Cu	Cd	Pb	As	Hg	Mn	Fe	SS	COD	A390
leachate	TM1	53	5.0	5.0	316	42	1.8	15	7500	280	920	3.08
	TM2	33	3.3	3.1	152	35	2.3	11	3250	790	725	2.95
	NS1	50	29	4.0	102	53	82	2200	10400	0.16	1010	2.31
surface water	TM3	7.8	22	1.4	92	17	3.6	60	9610	170	43.4	0.15
	NS2	13	8.8	2.0	27	37	420	3200	18400	0.14	535.6	0.83
	NS3	12	31	2.0	21	31	60	ND	14100	0.22	150.1	0.29
VN standard for surface water B		1000	1000	20	100	100	2	800	2000	80	35	—
VN standard for industrial water C		2000	5000	500	1000	500	5	5000	10000	200	400	—

Unit: Metals; ppb, SS and COD; ppm

RESULTS AND DISCUSSION

Characteristics of leachate and surface water collected from Tay Mo and Nam Son waste landfill site

Tables 1 shows the quality of leachate and surface water collected from Tay Mo and Nam Son waste landfill sites. Leachate samples, TM1 and TM2, collected from Tay Mo landfill, showed significantly high COD, SS and A390 value, which indicates the heavy color mainly derived from humic substance. TM1, TM2 and the surface water sample, TM3, collected from the Nhue River, which runs nearby Tay Mo landfill, contained extremely high concentration of mercury and iron and the concentrations of mercury and iron exceeded Vietnamese standard for surface water B. It was suggested that Tay Mo landfill still had a potential for environmental pollution though it passed six months since it had been closed. Leachate sample, NS1, collected from Nam Son landfill and the surface water samples, NS2 and NS3, collected from the reservoirs which were located around Nam Son landfill, showed high concentrations of COD, mercury and iron. The concentration of Manganese in NS2 was over the limitation of Vietnamese standard for surface water B. Although high concentration of lead and arsenic were detected in NS1, the concentrations were under the Vietnamese standard for industrial water C. Amongst these 3 samples, NS1 showed the highest A390 value.
Jansen and Christensen (1999) had reported that 600-22000 ppb of iron and 250-2000 ppb of manganese were detected from four Danish landfill leachate. The concentration of iron in waste landfill leachates reported by Krueger *et al.* (1989) was less than 200 ppb and lower than the results obtained from this study. Yasuhara *et al.* (1997) reported that arsenic was detected from three out of eight Japanese landfill leachates and the maximum concentration was 27 ppb. However, a few report have described about arsenic and mercury in leachate. These results indicated that leachates investigated in this study were highly polluted by heavy metals compared to leachates previously reported. Furthermore, according to the review by Lema *et al.* (1988), the leachate COD values obtained in this study were higher than the range of the representative data of the landfill leachates in developed countries such as Netherlands, UK, USA, Canada, Spain and France, though the COD of Greek landfill leachate ranged between 3812-6489 ppm (Fatta *et al.*, 1998).

COD and color removal from leachate by AOPs

Figs. 1 and 2 show the COD and color removal from leachate collected from Tay Mo waste landfill by several AOP systems. Fenton (pH 3) and photo-Fenton (pH 3) were effective for removal of both COD and color. The removal efficiencies of COD and color by these two processes were almost the same extent, COD removal was 75 % and decolorization was 85 %. It has been reported that photo-Fenton reaction usually generates hydroxyl radicals much more than Fenton reaction, because ferric iron is regenerated via photoreduction of ferrous iron (Pignatello, 1992). However the effects of photo-Fenton reaction on COD removal and decolorization were almost the same extent with Fenton reaction in this study. It was suggested that heavy black color of leachate inhibited the penetration of UV light to the whole reactor. From the practical point of view, Fenton reaction would be effective enough for COD and color removal from leachate. And the optimization of UV irradiation should be indispensable when it was applied to the treatment of highly colored wastewater.

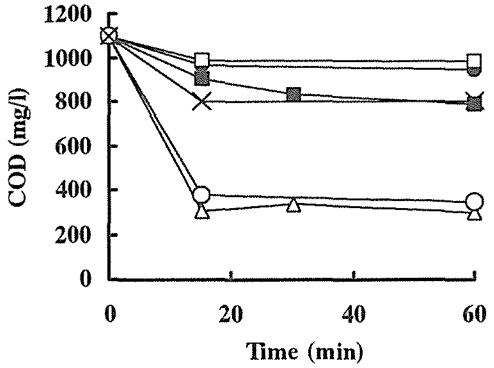

Fig. 1 COD removal from leachate by AOP treatment. □:UV-TiO_2, ●: UV-TiO_2-H_2O_2, ■:Fenton, △:Fenton (pH=3), ×:UV-Fenton, ○:photo-Fenton (pH=3)

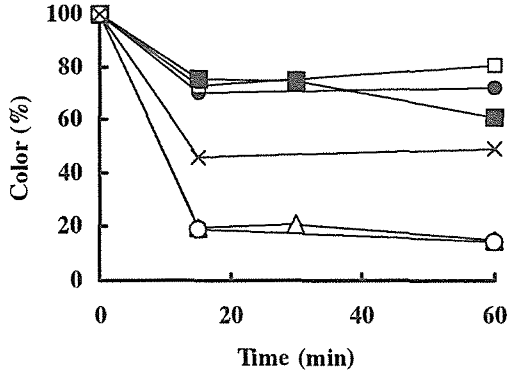

Fig. 2 Decolorization of leachate by AOP treatment. □:UV-TiO_2, ●: UV-TiO_2-H_2O_2, ■:Fenton, △:Fenton (pH=3), ×:UV-Fenton, ○:UV-Fenton (pH=3)

Fig. 3 shows the distribution of molecular weight of humic substances in the raw and AOP treated leachates. Distribution patterns showed no differences in each processes of AOPs, and the size of peak around the 60 ml and 150 ml of elution volume decreased uniformly. It was suggested the nonspecific and homogenous oxidation of humic substances occurred in AOPs. Furthermore, decolorization of leachate would be ascribed to degradation of humic substances with high-molecular weight.

Combination of AOPs and coagulation

The COD and color removal from the leachate by combined treatment of AOPs and coagulation are shown in Fig. 4. Coagulation of raw leachate resulted in removal of 24.2 % of COD and 3.6 % of color. The combination of coagulation and an AOP slightly improved the COD removal efficiency. However the effects of the combination were lower than the expected results which was estimated from the results of coagulation and AOPs in single unit operations. It was suggested that AOPs preliminary degraded organic compounds which might be removed by coagulation. In contrast, decolorization by coagulation was improved by combination with UV-TiO_2-H_2O_2 reaction and 45.3% of decolorization was achieved. These results indicated that the combination of an AOP and coagulation did not show remarkable efficiency compared to the single reaction of Fenton (pH 3) or photo-Fenton (pH 3), therefore, it can be said that combination of coagulation and an AOP would not be a sufficient leachate treatment process. Coagulation of leachate after Fenton and photo-Fenton

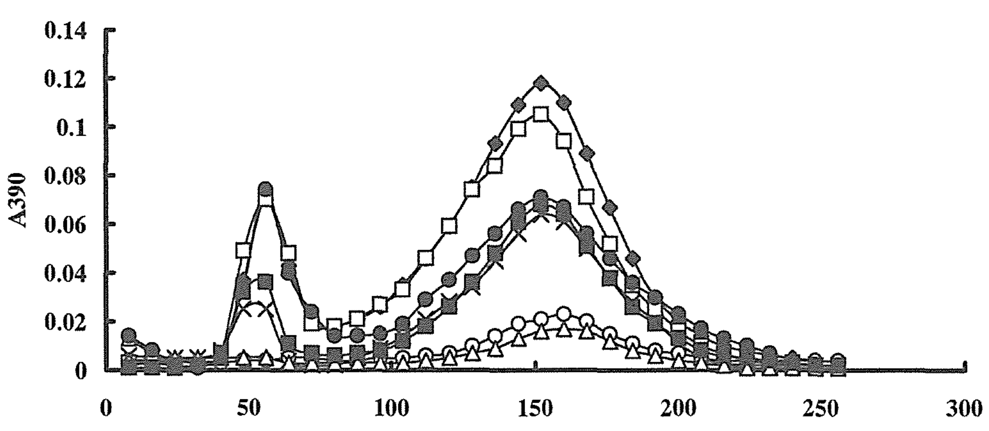

Fig. 3 Distribution of molecular weight of humic substances in leachate. ◆:raw leachate □:UV-TiO_2, ●: UV-TiO_2-H_2O_2, ■:Fenton, △:Fenton (pH=3), ×:photo-Fenton, ○:photo-Fenton (pH=3)

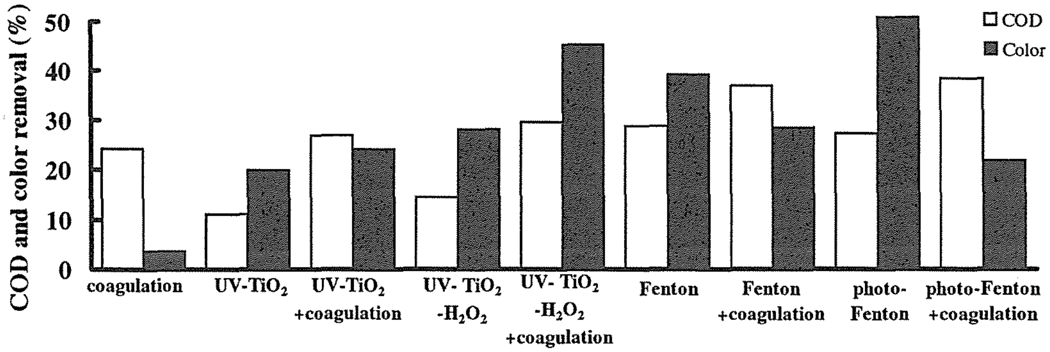

Fig. 4 Removal of COD and color by combination of AOP and coagulation

reaction slightly improved the COD removal, whereas their color was increased again. In the coagulation process, ferric iron would be converted to the ferric hydroxide and was then precipitated. It has been known that a large amount of addition of inorganic coagulants disperse the flocculation because of the result of positive charge of floc. Regarding with this result optimal of addition of ferric iron should be considered.

Ecotoxicological assay of leachate

Fig. 5 shows the results of *umu* test which evaluated the mutagenicity as a ratio of β-galactosidase activity of leachate to that of negative control. Although mutagenicity was not detected from raw leachate, the leachates treated by UV-TiO_2-H_2O_2, Fenton (pH 3) and photo-Fenton (pH 3) showed the significant level of mutagenicity. In contrast, estrogenic activities of all leachates were negligible. Since the assays adopted in this research were merely preliminary screening tests, raw leachate could not be concluded as non-mutagenic or non-estrogenic. However it was clear that three processes, UV-TiO_2-H_2O_2 process, Fenton (pH 3) and photo-Fenton (pH 3), increased mutagenicity of the leachate. It has been reported the unintentional production of mutagenic by-products during the degradation of organic compounds in AOPs (Manilal *et al.* 1992). In this study, organic compounds had still remained in the leachate after the treatment, suggesting the accumulation of such toxic by-products were produced through complicated degradation and synthesis in the AOPs. In order to manage the environmental risk, the optimum operating condition of AOPs, such as amount of chemical addition and/or reaction time, should be investigated to control the toxicity of leachates.

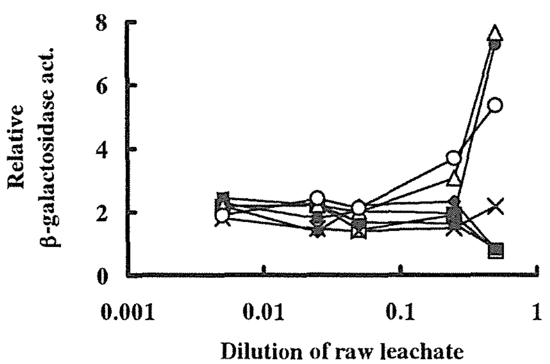

Fig. 5 Distribution of molecular weight of humic substances in leachate. ◆: Raw leachate, □:UV-TiO_2, ●: UV-TiO_2-H_2O_2, ■:Fenton, △:Fenton (pH=3), ×:photo-Fenton, ○:photo-Fenton (pH=3)

CONCLUSION

The leachates collected from waste landfills, which have received the industrial and municipal waste in Hanoi, showed the high concentration of heavy metals such as lead, iron, mercury and arsenic. High strength of COD and color were also shown in the leachates. The application of TiO_2-UV, TiO_2-UV-H_2O_2, Fenton reaction (with and without pH adjustment to 3) and photo-Fenton reaction (with and without pH adjustment to 3) revealed that Fenton (pH 3) and photo-Fenton (pH 3) processes effectively removed COD and color from Tay Mo landfill leachate. These processes were also capable for degrading the humic substance in the leachate. In contrast, combination of an AOP and coagulation processes did not show good results compared to the single reaction of Fenton (pH 3) or photo-Fenton (pH 3). Mutagenicity of leachate was increased by TiO_2-UV-H_2O_2, Fenton (pH 3) and photo-Fenton (pH 3) reaction. It suggested that degradation of organic matters in leachate resulted in generation of toxic intermediates. Selection of suitable AOP for leachate treatment must be carefully done from several viewpoints such as treatment efficiency, running cost, environmental toxicity and so on. This study could give an useful and practical information for grasping and improving the situation of Vietnamese waste landfill management.

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