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STUDY ON TREATMENT OF THE LEACHATE FROM LANDFILL SITE AT NAMSON, SOCSON, HANOI

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ABSTRACT

Landfill leachate has been treated by two step biological technology and advanced techniques. It was proved experimentally that biologically treated landfill leachate can not meet demand of existing Vietnamese standards for industrial liquid discharge. Application of Fenton reagent in combination with PAC adsorption can solve this problem. The application of PAC can increase COD removal efficiency of a unit H₂O₂ applied as much as twice or higher in comparison with reference data. Biologically treated or raw leachate can be filtered by RO technique. COD, colour and TDS removal were extremely good, but N-compounds removal was still poor. The best way to treat leachate to meet Vietnamese standards 5945-1995 B is to combine biological treatment with either oxidation-adsorption technique or with RO filtration.

Keywords: adsorption, advanced oxidation, Biological treatment, Fenton reaction, Landfill leachate.

Introduction

In the past, all over Vietnam the only technique applied for treatment of municipal solid waste (MSW) was dumping and simple landfill. Since 1998, the first constructed sanitary landfill was constructed in Namson site, Socson, Hanoi. This site, which has an area of 80 ha, is located 50 km Northern the Hanoi City and was planned to receive as much as 1,200 ton MSW per day. Along with landfill cells for MSW, a waste water treatment facility (WTF) for collected leachate consisting of two steps of biological treatment sequence was constructed too. The block scheme of this WTF is described as following:

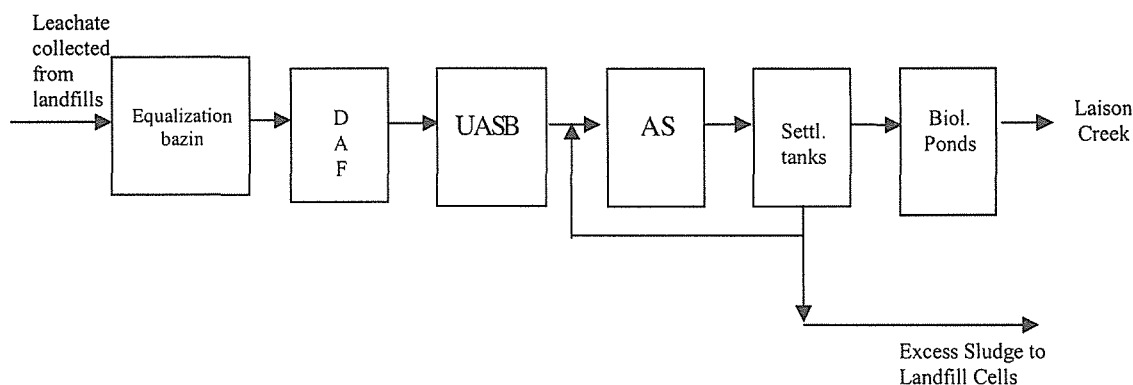


Figure 1. Flow diagram of existing leachate treatment facility at Namson Landfill site

The existing WTF has 8 UASB reactors operating simultaneously, connected with 4 parallel aerated tanks and 4 parallel settling tanks, which forms a treatment sequence: anaerobic-aerobic. The hydraulic detention times at UASB reactors and at aerated tanks were 12 and 8 hours respectively.

In 2000 this facility was started-up. During the trial of this system it was revealed that influent COD was low (up to 1,300 mg/L), that meant the leachate was “old”; influent pH was always higher than 7 meant anaerobic

acidification phase was accomplished and as a rule of these two features low BOD₅/COD ratio of about 0.2 and persistent to biological treatment characteristic of the leachate were observed: the treatment efficiency could not exceed 50% regarding to influent's COD. By the time, according to Vietnamese standards 5945-1995 for industrial wastewaters effluent COD has to get at least the COD value of 100 mg/L. The target of this paper is to find the way of upgrading /or retrofitting existing WTF so that it could get Vietnamese standards.

Land fill leachate characteristics and discharge requirements

It is well known that no two landfills produce the same quality of leachate and the quality of leachate strongly depends on many factors such as MSW composition, the way of management of landfill, the "age" of leachate and regional climatologic patterns [C. D. Martin et al. 1999]. At present in Namson landfill site, due to the lack of separation facility of the garbage the last one has complicated origin, including kitchen waste (mostly organics of natural origin), paper, plastics and even household chemicals, industrial wastes etc. That's why it is expected that many compounds toxic to human, recalcitrant and inhibitive to biological processes applied in wastewater treatment technology can be found in the leachate. Therefore, the composition of the leachate is very complicated and it is not easy to determine quantitatively each organic compound alone, therefore commonly, we have to be satisfied with the global parameters such as COD and BOD₅, which express indirectly the amount of organics present in wastewater and the part of organics being treated by microorganisms (in biological treatment) respectively. The other important feature of the leachate is its age: the younger the leachate, the higher COD, BOD values and BOD/COD ratio, the lower ammonia content. In contrary, the older the leachate, the lower COD, BOD values and BOD/COD ratio (that means more difficult to be treated by biological processes), the higher ammonia content (table 1).

Table 1. The content of Namson leachate

Items	"Young"	"Old"	VN Standard 5945-1995		
			A	B	C
pH	6-7	7-8	6-9	5.5-9	5-9
BOD ₅	3 000-20 000	250-350	20	50	100
COD	5 000-36 000	1000-1400	50	100	400
TSS	200-500	200-300	50	100	200
TNK		200-250	30	60	60
PO ₄ ³⁻		20-25	4	6	8
Coliform, MPN/100mL		50 000	5 000	10 000	

Note: All figures are in mg/L unit, except pH and Coliform; Standard A- can be discharged into all receiving water bodies; B- can be discharged into receiving water bodies used for agriculture and fishery only; C- must be treated.

Table 2. International Discharge Standards of Leachate

Items	USA	Japan	UK	Germany	Switzerland	Australia	VN-B	
BOD ₅	100	160	-	60	20	20	25	50
COD _{Mn}	-	-	160	-	-	-	-	-
COD _{Cr}	-	-	-	-	200	-	90	100
SS	-	200	200	100	20	20	30	100
NH ₃ -N	35	-	-	80	50	-	-	1
			(NH ₄ -N)	(NH ₄ -N)				
NO ₂ -N	-	-	-	-	10	0.3	1.5	-
TKN	-	-	-	-	-	-	-	60
Year	1979	-	-	1989	1988	1981	1995	

By comparison to other International Discharge Standards of leachate (table 2) it was found that VN Standard 5945-1995 B may be one of the most strict.

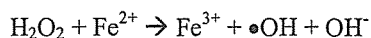
Our observation indicated that Namson leachate is referred to as “old” leachate, that’s why application of above mentioned two stage biological treatment lonely could not get demands of VN Standard B. The limitation of biological treatment was due to presence of large amount of recalcitrant organic compounds, some of them have high molecular weight, eg. humic substances [Ishigaki T. et al., 2001]. In this case, application of advanced oxidation processes, eg. Fenton reaction, could be a good option. There are some benefits of using Fenton reagent: 1- it is simplest way to produce •OH radical, a key constituent in AOP; 2- raw materials (FeSO₄ and H₂O₂) are available in the market and cheap; and 3- no need in complicated reaction devices.

It is the reason why most attention in this study was paid on combined physico-chemical treatment where the advanced oxidation process plays central role.

Advanced Oxidation Processes-AOPs

AOPs are processes that use *in-situ* formed •OH radicals with very high oxidation potential ($E_0 = 2.8$ V) for oxidation of contaminants.

There is a lot of systems which can produce •OH radical, eg. O₃/UV; H₂O₂/UV; O₃/H₂O₂; TiO₂/UV etc. Among them the oldest and may be simplest is Fenton reagent [Fenton, 1894]. Fenton reagent is the mixture of Fe²⁺ and H₂O₂. At pH 3-4 the production of •OH radicals occurred by reaction:



The main drawback of Fenton system is formation of sludge in the form of Fe(OH)₃ during neutralization reaction mixture. At present due to high oxidative and degradable potential of •OH radicals AOPs became interest of many researchers who is finding the way to treat recalcitrant compounds/or at least to break them down into low molecular weight compounds which can easier be treated biologically [Geenes et al., 2000].

Experimental and analytical procedures

Coagulation/or adsorption and Fenton oxidation experiments were conducted using 6-blades Flocclab (Prolabor-France).

Nitrification was conducted in batch mode, reactor, carrier material and procedure were described else where [Cao The Ha et al., 2001].

Reversed osmosis filtration was conducted using RO apparatus HA 3110 with capacity of 900 L/d, kindly provided by Toyobo Eng. Co., Ltd. (Japan).

Raw leachates were taken from Namson landfill site. There were two types of leachate: leachate No2, which was taken from collection well, represents leachate before biological treatment with the following characteristics: pH = 8.0-8.2; COD_{Cr} = 1052 mg/L; BOD₅ = 200 mg/L; A₃₉₀ = 2.290 and leachate No3, which represents leachate after biological treatment with the following characteristics: pH = 8.0-8.3; COD_{Cr} = 610 mg/L; BOD₅ = 10 mg/L; A₃₉₀ = 1.520.

All chemicals used in treatment experiments were technical grade. Powder activated carbon (PAC) was Trabac WT3 product. Flocculant was of Organo (Japan) polyacrylamide-PAA product N101 (nonionic). pH value was measured by Hanna pH Meter HI8314. Chemical oxygen demand (COD) was measured as dichromate oxygen demand (COD_{Cr}) according to standard methods (APHA, 1995). BOD₅-was measured by WTW 602 (Germany) according to standard method. NH₄-N was determined by indophenol method,

absorption was measured at $\lambda = 640 \text{ nm}$. Colour reduction was determined via reduction of Absorption value at $\lambda = 390 \text{ nm}$.

Results and discussion

Influence of pH value and order of chemical addition

pH values of the leachate were adjusted by addition of dilute solution of H_2SO_4 to desired values. In all experiments, except when desired to change, the concentrations of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ and H_2O_2 added were 2.45 mmol/L (680 mg/L) and 25.24 mmol/L (858 mg/L) respectively. The order of chemical addition was: first solution of H_2SO_4 for pH adjustment, second solution of Fe, third solution of H_2O_2 . The difference was only between first and second rows, when H_2O_2 was added after and before Fe (R-denotes reversed order).

Table 3. The influence of initial pH values of leachate and the order of chemical addition on effluent's COD

PH	7	6	5	4	3
Leachate No3 (COD _o = 610 mg/L)	-	600	565	170	291
Leachate No3-R (COD _o = 610)	-	557	549	356	273
Leachate No2 (COD _o = 1052)	713	699	620	225	270

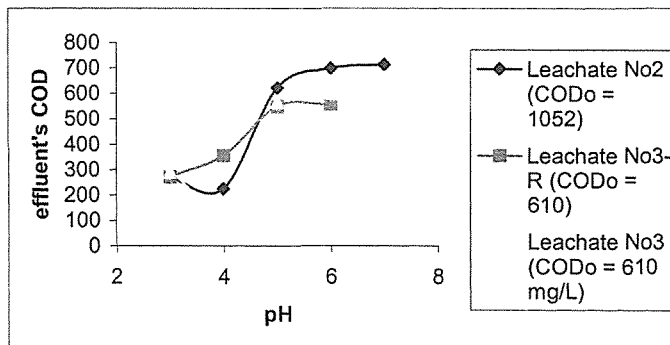


Fig 2. The influence of initial pH values of leachate and the order of chemical addition on effluent's COD

It was found from table 3 and figure 1 that in all cases pH of 3-4 is the best for Fenton reaction and reversed order of chemical addition (H_2O_2 was added before Fe) caused adverse effect. From now on the following best order of chemical addition was applied for all experiments: 1- pH adjustment by H_2SO_4 addition; Addition of FeSO_4 ; 3- Addition of H_2O_2 .

The role of PAC and PAA

Among all above mentioned series of experiments no result could get demand of VN Standard B. To improve effluent's quality an amount of PAC was added just after H_2O_2 addition (preliminary experiments on adsorption by PAC addition into settled effluent showed little effect). To enhance settling characteristic of treated leachate an amount of 1 mg/L of PAA was added after neutralization by adding NaOH to pH7. The order of chemical addition was as the best case as discussed above. The results obtained with PAC dose of 0.2 g/L were shown in tables 4a and 4b.

Table 4a. The influence of PAC doses on effluent's COD

PH	6	5	4	3
Leachate No3 (COD _o = 610 mg/L)	600	565	170	291
Leachate No3 + PAC 0.2 g/L	370	253	107	118

Results from table 4a showed significant positive effect of PAC adsorption on COD removal at all pH range. To clarify the effect of PAC adsorption a series of experiment at pH of 4 was conducted. Results were given in table 4b.

Table 4b. The influence of PAC doses on effluent's COD

PAC dose, g/L	0	0.16	0.20	0.24	0.28	0.32
Leachate No3 (COD _o = 610 mg/L)	173	120	107	106	103	100
Leachate No2 (COD _o = 1052)			220			

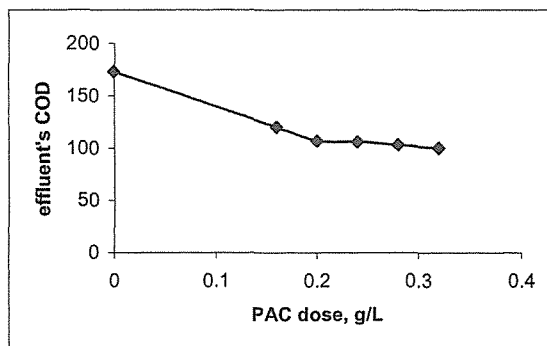


Fig 3. The influence of PAC doses on effluent's COD

Through tables 4a and 4b and figure 3, one can note that adsorption by PAC and flocculation by PAA had significant effect at the PAC doses of lesser than 0.2 g/L, in this range COD removal rate follows linear dependence on PAC doses, further increase of PAC dose continues to reduce COD but with lower rate. In the case of more strength leachate No2 the effect of PAC was not so significant, it may due to the fast saturation of PAC adsorption capacity by high concentration of organics in the initial leachate.

The role of H₂O₂ doses

The initial value of pH was kept constant and equals to 4 for all experiments. The only variable was H₂O₂ dose. The results obtained were shown in tables 5a, 5b and fig.3.

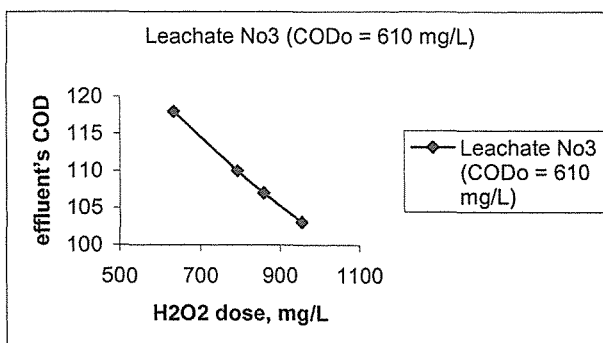


Fig 4. The influence of H₂O₂ doses on effluent's COD-Leachate No. 3

Table 5a. The influence of H₂O₂ doses on effluent's COD-Leachate No3

H ₂ O ₂ dose, mg/L	634	792	858	954
Leachate No. 3 (COD ₀ = 610 mg/L)	118	110	107	103

From table 5a one could calculate the COD removal capacity of H₂O₂:

$$\text{Removal} = \text{g COD removed/g H}_2\text{O}_2 \text{ applied} = [610 - (\text{eg. } 103)]/954 = 0,53 \text{ g COD/1 g H}_2\text{O}_2.$$

According to the results from the work of Lee [Jong-Yun Lee, 2001] this value was only of 0.2-0.3 g COD/1g H₂O₂, that means that our treatment has as much as twice higher COD removal efficiency accounted to the mass unit of H₂O₂ consumed.

For comparison purpose, the same experiments were done with leachate No2 with exception of not using PAC and PAA. Results were given in table 5b.

Table 5b. The influence of H₂O₂ doses on effluent's COD-Leachate No2

H ₂ O ₂ dose, mg/L	264	396	528	660	792	858	924
Leachate No2 (COD ₀ = 1052 mg/L)	304	297	263	258	234	231	225

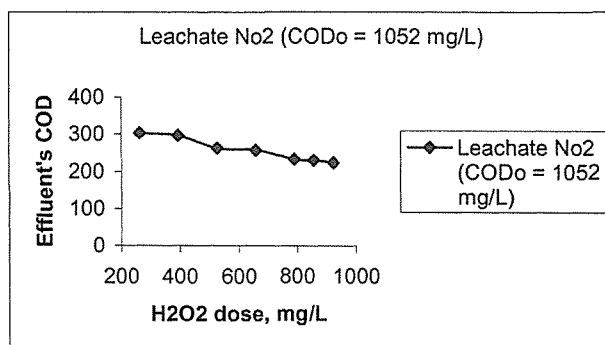


Fig 5. The influence of H₂O₂ doses on effluent's COD-Leachate No2

In this case:

$$\text{Removal} = \text{g COD removed/g H}_2\text{O}_2 \text{ applied} = [1052 - (\text{eg. } 225)]/924 = 0.895 \text{ g COD/1 g H}_2\text{O}_2.$$

Herein results were much better than that was in the case of table 5a and in [Jong-Yun Lee, 2001]. This phenomena may be explained by higher concentration of organics-substrate, in this case •OH radicals have higher probability of collision with substrate molecules than with abundant in water inhibitive ions such as bicarbonate.

The role of FeSO₄ doses

The initial value of pH was kept constant and equals to 4 for all experiments. The order of chemical addition was as the best case as discussed above. The only variable was FeSO₄ dose. The results obtained were shown in tables 6 and fig.6.

Table 6. The influence of FeSO₄ doses on effluent's COD-Leachate No3

FeSO ₄ doses, mg/L	408	544	680	816	952
H ₂ O ₂ : FeSO ₄ (mol : mol)	17.37	13.0	10.42	8.69	7.47
Leachate No3 (COD _o = 610 mg/L)	378	193	109	103	126

Obtained results in fig. 6 revealed the best case when H₂O₂ : FeSO₄ ratio equals to 8.7, this value is lower a bit in comparison with literature ratio of 10.

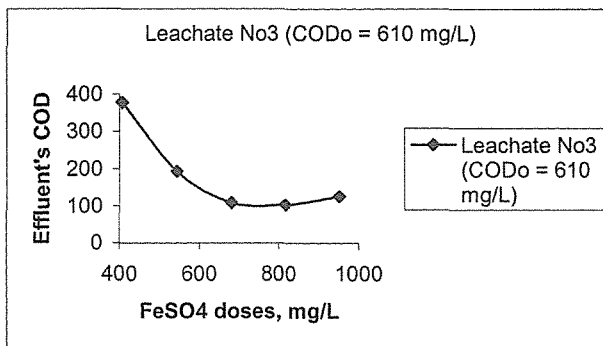


Fig 6. The influence of FeSO₄ doses on effluent's COD-Leachate No.3

Results of nitrification in batch mode

Nitrification was conducted in the batch with 20 L of carrier material from porous basal [Cao The Ha et al., 2001] to check the possibility of biological nitrification of the leachate. The results were given in table 7.

Table 7. Results of nitrification in batch

t, h	0	4	8	12	20	24	28	32	40
NH ₄ ⁺	152	118	82	52	22	10	7.5	4.4	0.9
NO ₂ ⁻	25	57	102	101	104	93	15	9	0.3
NO ₃ ⁻	91	166	234	367	423	486	597	627	642
COD	193	139	136	118	95				

Note: All figures are in mg/l

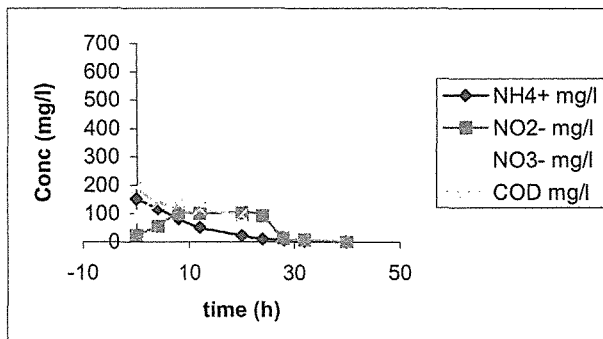


Fig 7. Results of nitrification in batch

These results confirmed the possibility of biological nitrification process. As a rule, denitrification can be accomplished much easier just supply enough BOD.

Results of RO filtration

RO filtration was conducted in the field. The results were given in table 8.

Results of RO filtration was excellent for both leachates, especially in regard to COD, colour and TDS removal. In the case of N-compounds RO appeared to be poor. To protect RO membrane from frequent clogging and consequence washing the RO column must be installed after pre-filtration unit.

Table 8. RO filtration results: before and after filtration

Leachate	BOD ₅	COD _{Cr}	TSS	TDS	TKN	NH ₄ -N
Leachate No2	250/10	1250/100	350/0	30,000/6,000	295/100	218/40
Leachate No3	50/-	650/8	200/0	25,000/5,000	120/40	100/40

Proposal for upgrading existing WTF

Based on experimental results of combined oxidation and adsorption experiments and RO filtration, the following scheme for treatment of Namson leachate was proposed (figure 8). This proposal includes the following main changes of existing WTF:

- Replacement DAF by coagulation-flocculation unit to remove SS, partly COD (most of all are high molecular weight fractions), partly Ca²⁺ which can cause scaling on all equipment and devices, especially on RO membranes.
- Conversion of parallel conventional aerated tanks into reactors in sequence for BOD, nitrification and denitrification. Including of the last two units is extremely useful in the case of application of RO filtration because of poor removal efficiency of RO process regarding to NH₄-N.
- There are two options for the last unit: either combined adsorption-Fenton oxidation or RO filtration can be applied. Selection of either Fenton oxidation or RO filtration should be made on the base of cost estimation. In the case of Fenton oxidation chemical cost will be the major part, in contrary in the case of RO filtration machinery cost is decisive. If ignore the cost, application of RO technique after biological steps promises more benefit in effluent quality and in less operational variables, especially in the case of very unstable influent quality of landfill leachate.

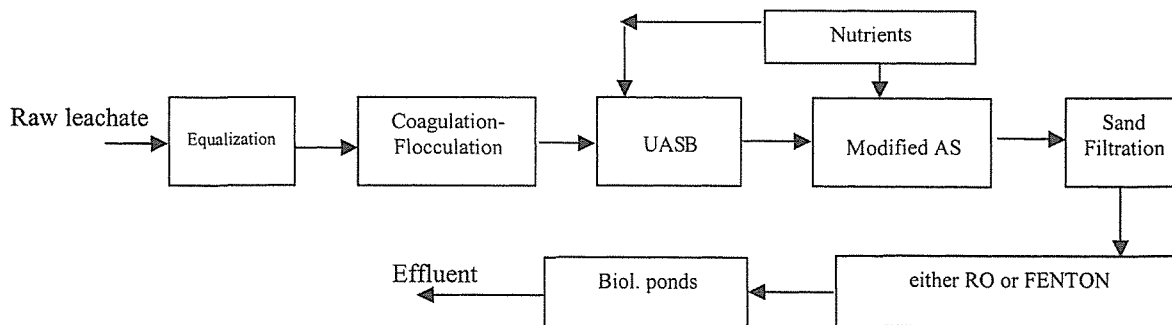


Figure 8. Proposed scheme for treatment of Namson landfill leachate

Conclusion

Due to high strength and complicated composition landfill leachate biologically lonely treated can not get demands of Vietnamese Standards 5945-1995 B.

To overcome this problem a combine approach must be considered. There are two options: after biological treatment to remove major part of organics and N-compounds following by either oxidation-adsorption process or RO filtration.

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