

Treatment of leachate by combining PAC and UV/O₃ processes

Kết hợp keo tụ với PAC và quá trình UV/O₃ để xử lý nước rỉ rác phát sinh từ bãi chôn lấp chất thải rắn

Research article

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The landfill leachate is commonly treated for non-biodegradable organic matters, ammonia and colour. Experimental investigations using polyaluminium chlorite (PAC) and UV/O₃ have been conducted for the determination of optimal pH value, reaction time and PAC concentration for the removal of chemical oxygen demand (COD) and colour. In pre-treatment coagulation stages, the highest COD and colour removal efficiencies were observed at the concentration of PAC $\geq 3,000$ mg/L and pH values between 7 and 8. However, these experiments also indicated significant removal efficiency for PAC starting with concentrations of 1,500 mg/L. The efficiency of COD and colour removal were approximately 30% and 70%, respectively. Similar efficiencies have been observed also during the second treatment stage where UV/O₃ processes were used to treat coagulated leachate. After UV/O₃ application, the pH of leachate reached the optimum value of 7.5 whereas the highest COD and colour removal efficiency was 55% and 72%, respectively, and the optimal reaction time was achieved after 80 min.

Nước rỉ rác sinh ra từ bãi chôn lấp chất thải rắn cần được xử lý các thành phần chất hữu cơ khó phân hủy sinh học, xử lý amoni và độ màu. Một số kết quả thử nghiệm về xử lý COD và màu của nước rỉ rác bằng việc sử dụng phương pháp keo tụ với PAC và quá trình UV/O₃ đã được thực hiện cùng với việc xác định các giá trị pH tối ưu, thời gian phản ứng và nồng độ PAC tối ưu. Hiệu suất xử lý cao nhất đạt được khi nồng độ của PAC ≥ 3.000 mg/L, pH trong khoảng từ 7 đến 8 trong giai đoạn tiền xử lý. Tuy nhiên, hiệu quả loại bỏ COD và màu bắt đầu tăng rõ khi nồng độ PAC từ 1.500 mg/L trở lên. Hiệu quả loại bỏ COD và màu tương ứng là khoảng 30% và 70%. Các giá trị pH này phù hợp cho quá trình phản ứng UV/O₃ được sử dụng sau giai đoạn keo tụ. Sau quá trình xử lý bằng hệ UV/O₃, pH của nước rỉ rác tối ưu được xác định là 7,5 (hiệu suất xử lý COD và màu cao nhất tương ứng là 55% và 72%), thời gian phản ứng tối ưu là 80 phút.

Keywords: landfill leachate, PAC, UV/O₃, COD, colour, ozone

1. Introduction

Currently, solid wastes generated in urban areas in Vietnam are not been treated thoroughly. Leachate generated from landfill sites is not controlled and treated properly. Burial is the common treatment option of municipal solid waste in Vietnam because this is a simple and low-cost technique. However, there are several jumble hazardous components such as batteries, engine oil, chemicals, toxic waste from household, commerce, and even industries, etc. that can emit heavy metals, toxic organic compounds into the air, water or ground. Today, landfill sites in Vietnam generate large amounts of leachate con-

taining highly toxic and non-biodegradable organic chemicals caused by the continuous decomposition of solid waste (Khanh, 2007). If leachate is not properly treated, it will continue to pollute surface and groundwater, causing serious environmental contamination. Chemical oxygen demand (COD) is the most difficult problem in leachate treatment since long-time halogenated organic compounds with high molecular weight can be generated and endanger the environment once they are discharged into soil and water bodies (Phuoc and Cuong, 2007).

The use of UV/O₃ process to treat landfill leachate will have high performance because ozone oxidizes strongly

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organic compounds. However, because of its high pollution levels, leachate needs to be pre-treated. Coagulation processes are used to reduce suspended solids, organic matters of wastewater through agglutination and settling out (Khanh, 2007; Phuoc and Cuong, 2007). These processes reduce part of the persistent organic matters in leachate. Coagulated leachate is then oxidized by ozone with UV light to break down the chemical links of organic matters in landfill leachate.

2. Materials and methods

2.1 Researching scale

Landfill leachate was taken from Da Mai site, Thai Nguyen and Nam Son site, Hanoi city, Vietnam. It was kept in plastic cans and stored at 4°C in the laboratory before treatment. Table 1 shows the characteristics of landfill leachate from Da Mai site.

Table 1. The characteristics of landfill leachate from Da Mai site

Parameters	Unit	Range
pH	-	7.5 - 8.3
COD	mg/G ¹	2,780 - 4,000
Colour	Pt-Co	1,250 - 1,500

2.2 Experiment 1: Pre-treatment by coagulation – flocculation processes

Coagulation was used to treat leachate in experiments with polyaluminium chlorite (PAC) used as flocculant. The suitable pH values and flocculant concentration were chosen for the pre-treatment. The pH value and COD concentrations were measured by Standard Methods (APHA, 1995). The colour was analysed by spectrum method with Pt-Co unit at 420 nm (APHA, 1995). Pre-treatment experiment of leachate was carried out with PAC. The sodium acrylates Acrylamic copolymer A110 was used as auxiliary flocculant.

The experiments aimed to investigate the effects of coagulant concentration and leachate pH on treatment efficiency, thereby determining the suitable pH and concentration for flocculation process and advantage for the next stage.



Figure 1. Jar-test equipment

The coagulation-flocculation experiments were performed in a conventional jar-test apparatus equipped with six backers (Figure 1) at room temperature ($20 \pm 2^\circ\text{C}$). The experimental procedure consisted of three subsequent stages: the initial rapid mixing stage of 3 min at 150 rpm and then adding auxiliary flocculants (A110) for the last minute of the stage; a slow mixing stage of 10 min at 50 rpm was following, then the final settling step for 30 - 60 min. The pH values of all samples were adjusted to the desired level by adding appropriate amounts of 4N sodium hydroxide or 4N sulphuric acid. After the settling period, the leachate was withdrawn from the beaker and analysed.

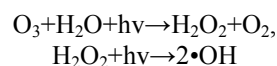
2.3 Experiment 2: Landfill leachate treatment by ozonation processes

After coagulation, leachate was treated by UV/O₃ system in the batch bubble reactor-type UV/O₃ apparatus (Figure 2) consisting of a mica column with a height of 100 cm and internal diameter of 5.4 cm. An UV lamp was added to improve oxidation of organic compounds by O₃. One litre of pre-treated leachate sample was then placed in the reactor while the sample's height amounted about 40 cm.



Figure 2. Schematic diagram of UV/O₃ apparatus

The ozone-air mixture was supplied from the bottom of batch reactor by ozone generator (model: Lin 4.10L) with nominal ozone output of 6.4 ghG¹. The airflow rate was 20 lmG¹. UV irradiation was used to activate the ozone molecules by absorbing the UV light with 39 Watt at 254 nm:



The experiments were designed to determine the optimal pH and reaction time.

3. Results and discussions

3.1 Pre-treatment by coagulation

3.1.1 Determination of suitable pH

The best conditions for coagulation - flocculation experiments were specified by considering COD and colour removal rates.

The effect of pH on COD removal

The effect of leachate pH value on COD removal efficiency by coagulation process is illustrated in Figure 3. Raw landfill leachate with high COD concentration is difficult to treat because it contains highly persistent organic matters. However, significant changes after flocculation process were observed in pH range from 3 to 10.

Figure 3 indicates COD concentration of raw leachate of about 2,643 mg/L, but the flocculation using PAC indicates that COD removal efficiency decreased with increasing pH value. The concentration of COD was reduced to 1,075 mg/L at pH = 4, equivalent of a removal efficiency of 59%.

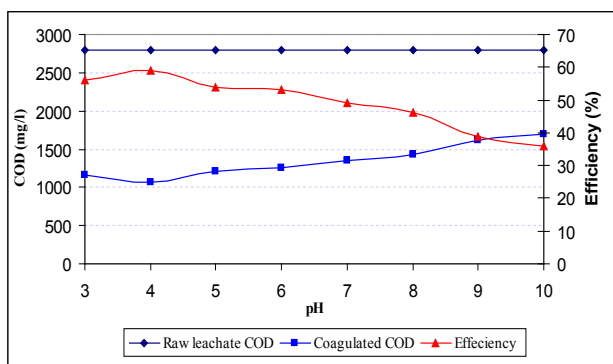


Figure 3. Effect of pH on COD removal by coagulation

The effects of pH on colour removal

The effect of landfill leachate pH on the change of colour in coagulation process is shown in Figure 4:

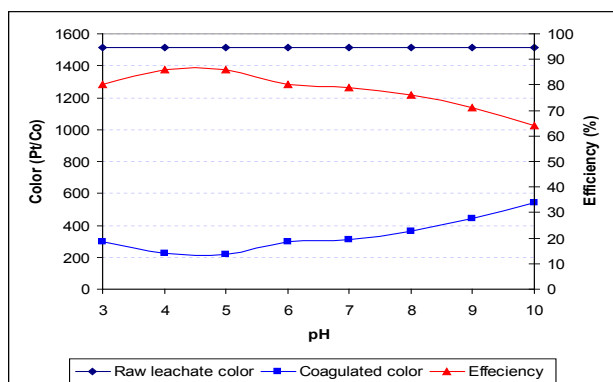


Figure 4. Effect of pH on color by coagulation

The efficiency of colour removal decreased with increasing the pH value of leachate. The most effective treatment was achieved at pH value around 4 (about 85%).

The removal efficiency was higher at low pH than at high pH because at low pH values (under 6), the humic and fulvic acids of landfill leachate were precipitated. COD and colour removal efficiency decreased with the increase of pH. At pH values of 6-7, COD and colour removal efficiencies were the same, while the colour intensity and COD concentration increased with pH of leachate from 8 to 10. This proved that coagulants are dissolved in alkaline environment.

Leachate was treated effectively in slightly alkaline and alkaline environment by UV/O₃ process, so pH from 7 to 8 was selected for further experiments to reduce the cost on chemicals for pH adjustment.

3.1.2 Determination of suitable PAC concentration

The pH range was determined to be about 7 to 8. The next experiments were conducted at this pH value while changing flocculants concentrations from 500 mg/L to 5,000 mg/L.

The effect of flocculants' concentration on COD removal

The effect of flocculants' concentration on treatment efficiency is illustrated in Figure 5.

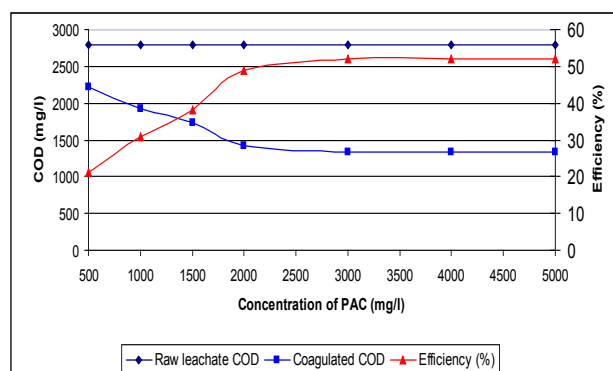


Figure 5. Effect of coagulants concentration on COD removal

Figure 5 shows that the performance of COD removal increases as the concentration of PAC increases (17 - 38%). COD decreased significantly after coagulation at the concentration of PAC from 1,500 mg/L. COD decreased from 2,798 mg/L to 1,340 mg/L while the concentration of flocculants increases from 3,000 mg/L or higher. In this case, highest efficiencies were achieved for all PAC concentrations.

The effect of concentration on colour removal

The colour removal rate by using coagulation increases quickly with flocculants' concentrations from 500 to 2,000 mg/L. The removal increases mostly in steps of 1,000 to 2,000 mg/L and then remains constant. The colour gradient went down quickly from 1,512 to 280 Pt-Co units (81%) at PAC concentrations of 2,000 mg/L and then was not significantly reduced. However, the colour intensity was still very high after flocculation.

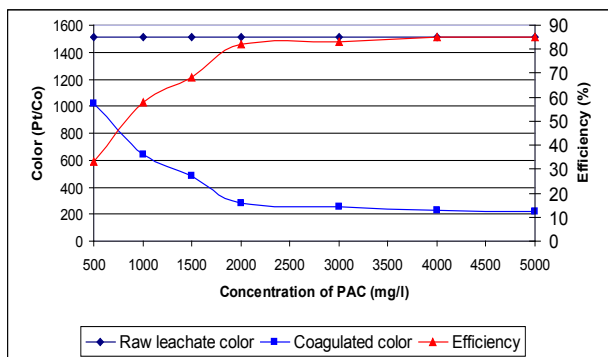


Figure 6. Effect of coagulant concentration on COD removal

3.2 Disposal of leachate using UV/O₃ process

3.2.1 Determination of optimal pH range for UV/O₃ process

The leachate was treated by coagulation with PAC and pH was not adjusted (after adding coagulants, pH dropped from 7.8 to 7.35) before treated by UV/O₃ process. Because concentration of COD of leachate of Nam Son was collected was about 20,000 mg/l, the concentration of PAC selected for pre-treatment was 2,500 mg/l. The UV/O₃ process was performed as follows: input airflow at 20 lpm, the reaction time was 40 min; the corresponding concentration of ozone generated is 4.27 g/l air. UV phototherapy (at 254 nm of wavelength and 39 watt bulb) was also used to combine with O₃ to treat landfill leachate after coagulation. The experiments were done with pH values varied from 5 to 10.

The effects of leachate pH on COD removal by UV/O₃ process

The results of investigations for the determination of the influence of leachate pH are represented in Figure 7. The data show that pH did not have an important effect on COD of leachate. However, the performance of COD removal was slightly higher in alkaline environment. In slightly alkaline reaction with UV light, ozone can react with water to form *OH radical which is a strong oxidant. The optimal performance was achieved at pH value \approx 7.5 (reaching 47% removal efficiency, COD decreased from 20,330 mg/l to 10,783 mg/l).

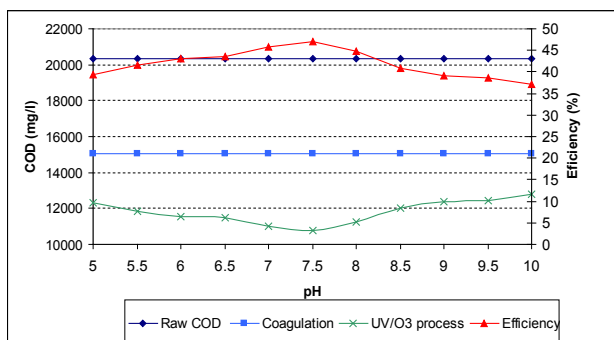


Figure 7. Effect of pH on COD by UV/O₃ process

The effect of leachate pH on colour removal by UV/O₃ process

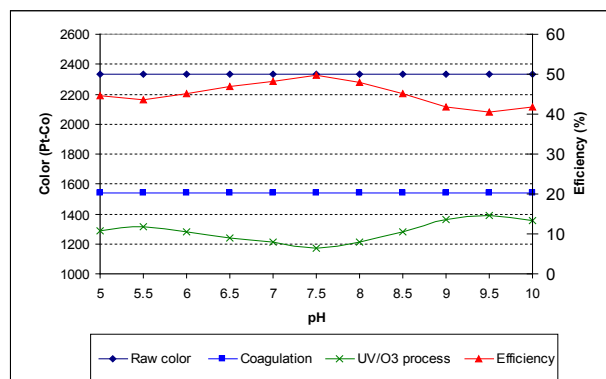
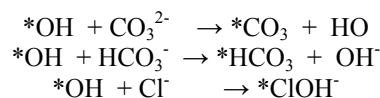


Figure 8. Effect of pH of leachate on color by UV/O₃ process

The experimental investigations have shown that the efficiency of colour removal was similar when changing the pH of the leachate. The removal efficiency was in general very high (from 41-50%) and took bigger values (45-50%) at pH values from 5 to 8.5 and at pH values from 9 to 10. When leachate pH was adjusted to 7.5, the efficiency of colour removal was highest after coagulation and UV/O₃ process. At high pH values, the anions Cl⁻, CO₃²⁻, HCO₃⁻ in leachate can react with the *OH radical generated by the UV/O₃ process:



Thus, removal efficiency of organic matters in leachate is reduced. The optimal efficiencies of COD and colour removal were achieved at pH: 7.5 so the pH value of leachate was set to 7.5 for the next experiments.

3.2.2 Determination of optimal reaction time

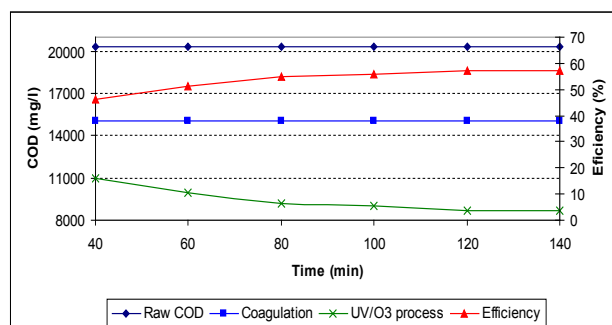


Figure 9. Effect of reaction time on COD removal

The experiments below were conducted using the following parameters: pH = 7.5, input airflow rate 20 lpm. The reaction time was ranging from 40-140 min and the corresponding ozone concentration was 4.27 g/l. The system was combined with UV bulb at all time during the treatment.

The effect of reaction time on COD removal

Figure 9 shows the reaction time for leachate treatment. The COD of raw leachate was 20,330 mg/l but it decreased to 15,051 mg/l with an efficiency of 26% after

flocculation by PAC. There was a significant reduction in COD after 40 min of reaction and the highest decline was corresponding to 9,207 mgL⁻¹ (by removing 55% COD) after 80 min after treating by UV/O₃. Then efficiency increased slightly when increasing the reaction time. In this case, the reaction of OH* with organic matters in leachate might have reached saturation levels.

The effect of reaction time on color removal

Similar to COD removal, the colour removal efficiency was high during the UV/O₃ process: the raw leachate was grey but it changed to black green after flocculation. The UV/O₃ process was conducted to treat leachate from 40 min to 140 min. It can be seen that leachate colour changed from black green to brown after 40 min of reaction, and then yellow in 60 minutes. After 80 minutes of reaction, the colour intensity was reduced to 647 Pt-Co units (removal efficiency of 72%). At 100 min the colour switched to white with 91% of colour removal. After 120 min, the efficiency of colour removal was 96%. As a result, the optimal reaction time was set after 80 min.

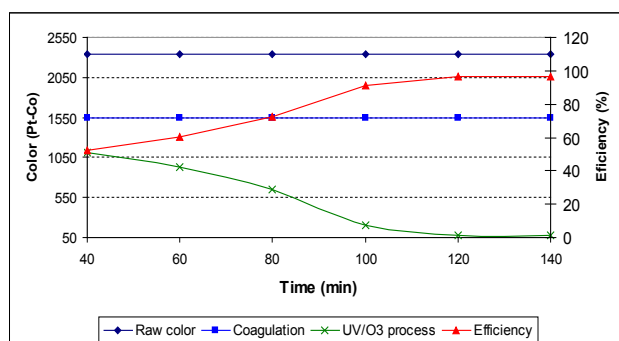


Figure 10. Effect of reaction time on color removal

4. Conclusion

Tests of coagulation of leachate showed that after processing using this method, the leachate met the standards for discharge. From the experimental phase, the following process-related parameters have been identified as optimum: PAC of 1,500 mgL⁻¹ at pH: 7-8. After UV/O₃ process, the optimal parameters determined are: pH = 7.5 and reaction time: 80 min. The process of combination of coagulation and UV/O₃ achieved high efficiencies: COD and colour removal of 55% and 72% respectively.

5. References

- [1] APHA 1995. Standard Methods for the Examination of Water and Wastewater. 19th American Public Health Association, Washington DC.
- [2] Khanh, N.H. 2007. Report of the project: Research and Comparison of Landfill Leachate Treatment Systems in Viet Nam and the World to Choose Suitable one to Meet Level B of TCVN for Landfill Sites of Hanoi city. Institute of Environmental Technology, Vietnam Academy of Science and Technology.
- [3] Phuoc, N.V., Cuong, V.C. 2007. Research to Increase the Treatment of Non-Biodegradable Organic Compounds of Landfill Leachate by Fenton Reaction. J. Sci. Scientific and Technological Development, Vietnam, 10: 71-78.
- [4] Phuoc, N.V., Phuong, N.T.T. 2006. Textbook of Treatment Techniques of Industrial Waste. Publisher of Construction, Vietnam, pp136-140.