Effect of pH on the Removal of Landfill Leachate Pollutant by DTRO

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Abstract. The operating pressure of the first-stage DTRO and the removal efficiency of COD, BOD, TP observed within 180 min at pH 6.5, pH 7, and pH 7.5, respectively, then the mechanism analysed and optimization methods recommended. The results show that the operating pressure reached its fastest increasing rate at pH 7.5, which grew from 32.13 to 32.66 Bar rapidly. The removal efficiency of all major pollutants increased steadily with uptime before stability. The maximum removal rates of COD, BOD, TP were 97.9%, 97.36%, and 97.7% at pH 7.5, respectively. The system performance can be improved by adjusting the pH value of the leachate to 6 ~ 7 during operation.

Keywords. landfill leachate; DTRO; pH; pollutant removal

1. Introduction

The effectiveness and operating conditions of the DTRO system for treating landfill leachate have been published in journals and conference papers [1-7]. However, few studies are available on how the pH value alter the working characteristics of RO membranes and thus affecting the removal efficiency of major pollutants. This article observes the removal efficiency of COD, BOD_5, and SS in a two-stage DTRO system under different pH values. The influence causes are analyzed, and the corresponding optimization methods are explored, for reference to other landfill leachate treatment projects.

2. Experiment and analysis

2.1 Materials and Methods

The landfill leachate treatment station is located in a plateau of Southwest China where belongs to the subtropical monsoon climate zone at an altitude of 1910m. The average annual temperature is 14.8 °C. The average minimum and maximum temperatures are 9.7 °C and 21.6 °C, respectively. The extreme temperatures reached -6.2 °C and 31.6 °C in the record. The average annual precipitation is 900mm, and the precipitation of rainy season (May-October) achieves about 86.7% of the total.
The experimental equipment includes a leachate storage tank, sand filter, precision filter, first stage DTRO, second stage DTRO, clean water tank, air compressor, pump, etc. The treatment scale is 100t/d, and the designed water production rate is 75%. The power for the system installed is 61.68kW, with an operating value of 37.35kW. The DTRO system test reagents mainly include concentrated sulfuric acid (H_2SO_4) and sodium hydroxide (NaOH) for pH adjustment, cleaning agent, and scale inhibitors for system cleaning.

2.2 Main pollutants of leachate

Table 1 listed seven major pollutants and five heavy metal ions detected in the landfill within a typical day.

Tab. 1 The main pollutants of influent (mg/L)

Pollutants	(mg/L)	Pollutants	(mg/L)
SS	121	Cr^{6+}	0.018
TP	36.5	Pb	0.264
COD	2.53×10^3	Cr	< 0.05
NH_3-N	203.3	Cd	0.008
BOD_5	434	As	0.118
TN	300.2		

2.3 Range of pH values

The main pollutants of leachate were detected from October to November considering stable climate and environment temperature in these two months. The sampling interval of each group was set to three or four days. Fourteen groups of data were collected during the testing period. Figure 1 significantly shows the pollutants removal efficiency such as COD correlated with the change of pH value.
The 13th and 14th records showed the removal efficiency changed greatly because of the hydraulic wash on landfill by rain showers. The operating pressure rose fast as a result of more SS (suspended solids) entered the DTRO. Higher osmotic pressure improves the pollutants removal. Besides the COD, and other main pollutants also show

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regular changes with the altering of pH value and operating pressure. The field experiments can immediately check the accurate effect of pH on the removal efficiency.

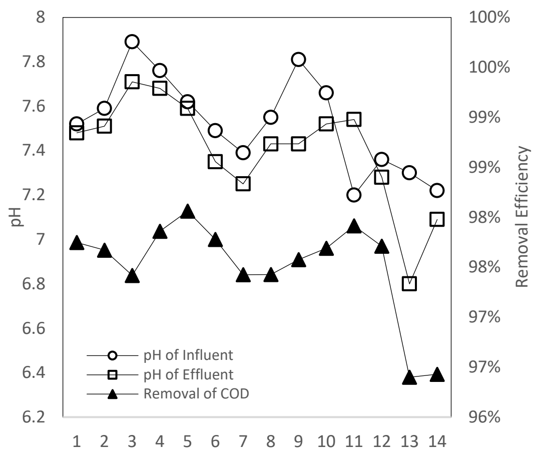

Figure 1. Relationship between pH and removal efficiency

The pH value of the influent recorded in 2 months was between 6.5 and 8. The pH drops after DTRO processing because of the RO membrane effects on acidic ions such as CO_3^{2-}, HCO^{3-} in liquid. As shown in formula 1, the CO_2 in the water produces a large amount of H^+ when it reacts to the right of the equation. Then it makes the effluent acidic as a result of $H^+CO_3^{2-}$ and HCO^{3-} cannot penetrate through the membrane and trapped in concentrated solution. The acidic effluent needs to be neutralized, considering the effluent would cause secondary pollution if the influent pH below 7.

$$CO_2 + H_2O \leftrightarrows HCO_3^- + H^+ \leftrightarrows CO_3^{2-} + 2H^+ \quad (1)$$

The carbonate, as a weak acid radical, is easy to combine with hydrogen ions to produce bicarbonate ions and hydroxide ions after ionization in water so that the concentration solution is alkaline. The operating pressure of DTRO increases rapidly as a result of the membrane fouling is aggravated when the pH value over 7. The economic and efficiency of DTRO would be reduced, considering more chemicals are required to deal with the high concentration pollutants and complicated components if the pH value is too high or too low. Therefore, it is appropriate to set the influent pH value to 6.5, 7, and 7.5 in the experiment, respectively. Then the effluent of DTRO was detected within 30, 60, 90, 120, 150, and 180 minutes.

3. Results and discussion

3.1 Effect of pH on operating pressure of the first-stage DTRO

Figure 2 illustrates the operating pressure rises steadily with the uptime. The highest-pressure was 32.66 bar at pH 7.5, 150 min. Additionally, the pressure reached 32.5 bar at pH 7 and 32.36 bar at pH 6.5, respectively.

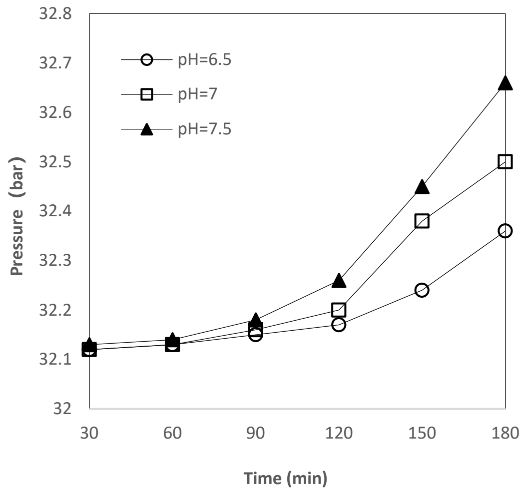

Figure. 2 Effects of pH on operating pressure

Alkaline leachate is likely to cause membrane fouling when pH over 7. The removal of COD, BOD, TP change regularly considering the operating pressure can directly affect the interception of pollutants. High pressure can improve the separation ability of RO to separate polymer materials, colloidal particles, and microorganism. But excessive pressure would make the salt transmission rate increases, then the removal efficiency gradually decreased.

3.2 Effect of pH on the removal efficiency of COD, BOD, and TP

Figures 3,4 and 5 show the trends of the removal of COD, BOD, and TP with the uptime. The change curves of these three pollutants seem similar under different pH value. The highest COD removal was 97.9% at pH 7.5, 180min. And the removal efficiency reaches 97.86% at pH 7 and 97.7% at pH 6.5, respectively.
The BOD_5 removal reached 97.36% at pH 7.5, 120 min, which was significantly higher than the value of 96.88% at pH 7, and 96.39% at pH 6.5, respectively. The removal efficiency showed a downward after 120 min.

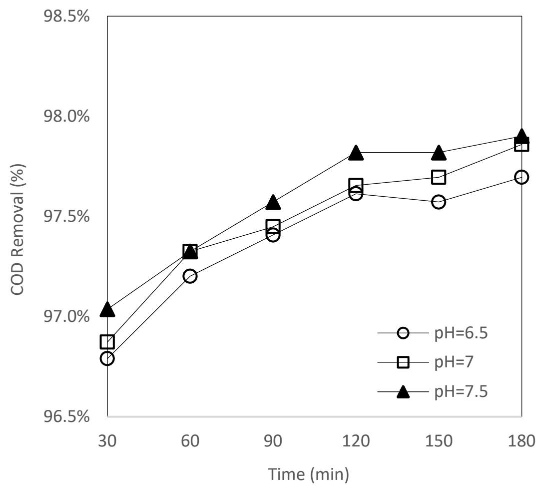

Figure. 3 Effects of pH on COD removal.

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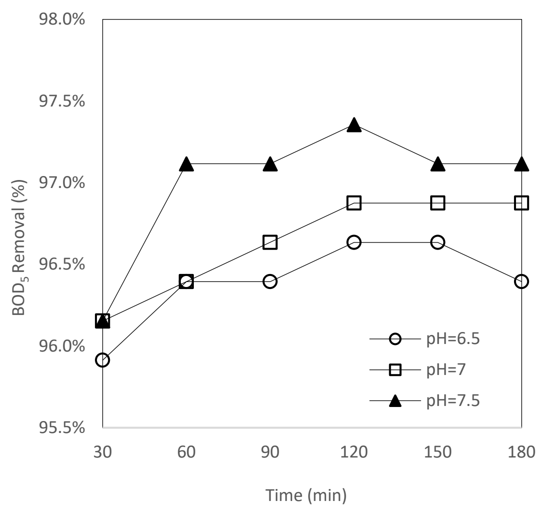

Figure. 4 Effects of pH on BOD5 removal

Figure 5 illustrates a more gradual change in TP removal. The removal efficiency at pH 7.5 is slightly higher than pH 7 and pH 6.5. The TP removal reaches the highest value of 97.7% at 150 min.

COD, BOD, and TP exist as various forms in the leachate. They may be suspended, colloidal, soluble, or be carried by organic matter and microorganisms. The leachate flows along the DT (Disk Tube) flow-path with a certain speed. The solvent, low molecular substances, and inorganic ions in the solution through the RO membrane enter the low-pressure side from the high-pressure side. RO can effectively intercept macromolecular substances and particles. Contaminants such as COD, BOD, and TP in the solution are trapped along with high molecular substances, colloidal particles, and microorganisms, which discharge as a concentrate.

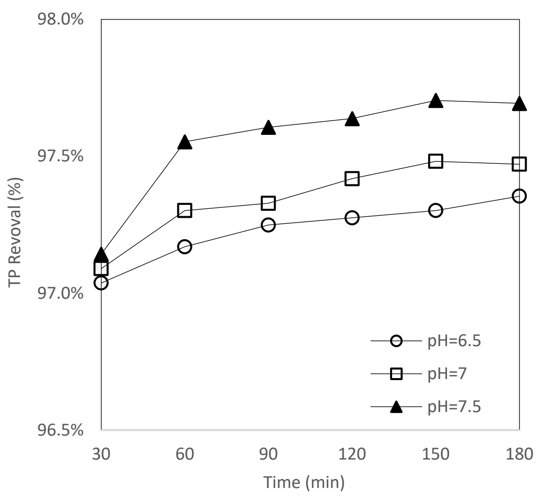

Figure. 5 Effects of pH on TP removal

The adsorption of polar organic on the surface of the RO membrane may be carried out by hydrogen bonding, dispersive force adsorption, and hydrophobic action [8]. Generally, there are three main influence factors of membrane effects on non-polar organics. First, the interaction between hydrophobic organics and water can enrich the slow-dissolving organics on the membrane surface. Second, the concentration polarization of polymeric organics also facilitates their adsorption on the membrane surface. Third, the interaction of ions in water (mainly Ca^{2+}) with organic functional groups alter the hydrophobicity and diffusivity of these organic molecules [9-11].

The pH affects the properties of the solute charge, and also the surface properties of the membrane. Thereby, the interaction between the solute and the membrane surface, the deposition amount of the solute and the membrane flux could be affected. Electro-adsorption on the surface of the RO membrane also effectively separates colloids and suspends solids from the leachate. The H^+ produced by the acidic leachate discharged through the RO membrane. Then a large amount of negatively charged ions are trapped by the membrane so that the membrane surface generates a repulsive force to the negatively charged colloid and macromolecular substance, which is not conducive to the adsorption of the surface [12]. Therefore, the adsorption performance changes and the removal efficiency increases when the pH value raised.

The Figures show that the removal efficiency generally increases or decreases slowly after 120 min when the concentration polarization reaches a certain value [13]. Because there still are a mass of vacancies on the surface for adsorption during the initial period. Differential concentration at the surface area caused by the rapid adsorption accelerates the diffusion of negatively charged particles and colloid. The vacant sites facilitate the rapid discharge of low molecular substances and inorganic ions through the RO membrane. However, the available adsorption sites gradually reduced with the uptime, and the passing ability decreased due to higher membrane surface coverage.

4. Conclusions

A large amount of charged ions were produced in the liquid when the pH value of the leachate changed. The accumulation of ions on the surface of the membrane contributes to the change of adsorption capacity. This is an important factor that affects the removal ability of the RO membrane.

Osmotic pressure also has a great influence on the separation efficiency of the RO membrane. Osmotic pressure accelerates the fouling phenomenon when pH over 7. The removal efficiency of the main pollutant is also improved when high osmotic pressure appears at the beginning. However, the pollutant removal efficiency does not rise but falls instead, due to concentrated polarization after the osmotic pressure reaches the critical point. Therefore, the pH value of leachate influent should be in the range of weak acidity. The weak acidic environment not only controls the net thrust of the system to a reasonable interval but also maintains stable pollutant removal efficiency.

The optimized pH value of the influent should between 6 and 7, which can decrease the dosage of chemicals to maintain better economics during operation, and obtain a better removal effect considering the proper operating pressure and adsorption capacity of the membrane surface.

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