Research of Landfill Leachate Treatment by Polymeric Ferric Silicate-Sulfate

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Abstract. In this paper, the treatment of landfill leachate by Polymeric Ferric Silicate-Sulfate was studied with the landfill leachate of Shankou landfill in Guilin city as the research object. The orthogonal experiment shows that the influence of dosage of PFSS, pH and dosage of PAM on the treatment is decreasing in turn. The removal rate of COD, TP, SS reaches at 71.09%, 99.13% and 97.54% when the original concentration of COD, TP, SS is 1442 mg/L, 7.94 mg/L and 244 mg/L respectively by setting the optimum parameters of PFSS 20 ml/L, PAM 20 mg/L, pH 6.5. Adding enough PFSS is beneficial to the rapid release of CO2 and the mud-water separation of flocculation, and avoid the influence of bubbles on the later separation of mud from water.

1. Introduction

The treatment of landfill leachate has always been an international problem. The leachate treatment processes in Shankou landfill in Liucheng county adopt traditional biological anaerobic aerobic, ultrafiltration and nanofiltration. However, due to the high concentration of pollutants in leachate and the large fluctuation of water quality and quantity, the subsequent membrane filtration is blocked seriously and the process is difficult to run.

Comparing with conventional flocculant, polysilicate metal salt which is a kind of inorganic high molecular flocculant has the advantages of simple preparation process, low price, high efficiency, small sludge volume, strong electric neutralization, and easy settling of bigger floc, good adsorption and bridge building and precipitation net trapping function and so on. By adding polysilicate ferric sulfate (PFSS) and organic flocculant (PAM), the suspended particles in the wastewater can be rapidly destabilized and aggregated into alum flocculus, which is beneficial to precipitation separation and subsequent membrane filtration, and effectively remove the pollutants such as organic matter, turbidity, bacteria salts and heavy metal ions in the wastewater[1-4]. With the development of ecological environment, municipal solid waste (MSW) landfill is becoming more and more widely. Flocculation process provides a useful method for the treatment of landfill leachate[5].

2. Water sample, reagents and experimental methods

The water sample was the raw waste leachate collected by the adjustment pond of Shankou landfill site in Guilin city, which was dark brown and accompanied by fetor. The water quality characteristics of leachate were tested as follows: pH 8.1~8.3, COD 1400~2000 mg/L, NH3-N 1400~1600 mg/L, TP 5~8 mg/L, SS 200~900 mg/L. The quality of the water sample required to be remeasured in each experiment, because of the storage time has great influence on the water quality of leachate.
The coagulant was the PFSS liquid produced by guangxi foster environmental protection co. LTD, with the density of 1.29 g/cm³, the SiO₂ quality score of 1.00% and the total iron quality score of 9.25% which we adopted. The flocculant was anionic polyacrylamide produced in Xilong chemical industry, with a specific grade and molecular weight of 3 million. The flocculant was configured into a solution of 1‰ concentration before use.

The experimental method was mainly carried out in four stages according to the process operation: (1) Coagulation stage, sample and add PFSS, and rapidly stir of 500 r/min for 15 min for coagulation reaction. (2) Adjust pH. (3) Flocculation and sedimentation stage, Adding the PAM solution while stirring, stirring at a moderate speed of 100 r/min for 1 min for mixture followed by stirring at a slow speed of 60 r/min for 2 min for flocculation, and settle for 30 min, measure the pH, COD, TP and SS of the treated water samples respectively.

3. Results and analysis

3.1. Effect of PFSS dosage on leachate treatment effect

In the coagulation stage, the dosage of PAM was 30 mg/L, and the pH value was 6.5. The experiment was carried out by changing the dosage of PFSS at 15-30 ml/L, as shown in figure 1.

As shown in figure 1, when the PFSS dosage increased from 15ml/L to 27ml/L, the COD removal rate increased from 61.69% to 78.18%. When the PFSS dosage was greater than 27ml/L, the COD removal rate was tending to be stable. TP removal rate was increased with the increasing of PFSS dosage, with removal rate higher than 96% and concentration lower than 0.25 mg/L, reaching the emission standard of domestic waste landfill pollution control standard (gb16899-2008).

When the dosage of PFSS increased from 15 mg/L to 21 mg/L, the removal rate of SS increased from 87.72% to that of 97.88%. As PFSS hydrolyzed in leachate, the ferric salt hydrolyzed compounds were adsorbed by the suspended particles containing humus in the water sample during the mixing process, and the particles were condensed after detaching from stable state. At the same time, Large flocs formed by bridging and adhesion of suspension of iron hydrolysate adsorbed by polysilicic acid macromolecules or sols⁶, humic colloids, Suspended solids and phosphorus compounds were removed through floc sedimentation. However, with the increase of PFSS dosage, the pH of the mixture gradually decreased from 6.54 to 2.80, and the flotation appeared in the flocculation process, and the removal rate of SS decreased rapidly to 40.27%. Too much bubble production seriously affected the flocculation and settling of sludge granules, so the appropriate dosage of PFSS was 19~21 mg/L.

3.2. Effect of pH on leachate treatment
pH value has a significant effect on the electrochemical environment and coagulation effect of leachate, and can strongly affect the treatment effect of PFSS on leachate\cite{7-8}. The dosages of PFSS and PAM were controlled as follows: 20 ml/L, 30 mg/L, and the pH value of the water sample was adjusted to 6.0–8.5. The leachate treatment effect is shown in figure 2.

![Figure 2. The removal effect of COD, TP and SS in leachate under different pH.](image)

From figure 2, it could be seen that the removal rates of COD and TP decreased gradually with the increase of pH, and the removal rate of COD decreased obviously from 70.16% to 56.28%, the removal rate of TP decreased from 98.98% to 93.65%. When pH was increased from 6.0 to 7.5, the removal rate of SS increased from 94.65% to 97.67%, but when pH was high beyond 7.5, the removal rate of SS began to decrease, and the removal rate of SS decreased to the lowest 88.37% when pH was 8.5. Because the hydrolytic products of Fe had little charge when pH was at the range of 8.0–8.5, the iron polymer produced by PFSS hydrolysis lost the ability of neutralization and destabilization, and the suspended pollutants such as humus colloid in leachate could not be effectively combined together to form floc and be removed. Neutral acid flocculation environment was more conducive to the flocculation treatment of leachate, which was consistent with previous research results\cite{5}.

3.3. Effect of PAM dosage on leachate treatment

PAM polymerizes the flocs formed by coagulation through the double interaction of electric neutralization and adsorption bridging. Under the action of shear force and centrifugal force of stirring device, the flocs are continuously collided to form large particles to realize the separation of mud and water. The experiment was carried out by controlling the dosage of PFSS for 20 ml/L, pH 6.5, and changing the dosage of PAM to 10 ~ 45 mg/L. The removal effect of COD, TP and SS in leachate are shown in figure 3.
Figure 3. The removal effect of COD, TP and SS in leachate under different dosage of PAM.

It was shown in figure 3 that both COD and TP can be removed effectively when the dosage of PAM was 10~45 mg/L. With the increase of PAM dosage, the removal rate of TP and COD was stable with the increasing of PAM dosage, the average removal rate of COD and TP was 70.40% and 98.78%, respectively, however, when the dosage of PAM was more than 30 mg/L, the removal rate of SS decreased. It was found that when the dosage of PAM was less than 20 mg/L, the flocs formed were smaller and looser, because of too little PAM dosage so that insufficient adsorption bridging. When the dosage of PAM was more than 30 mg/L, there was PAM residue in the water sample. When PAM was added in excess, colloid restability would occur, which would lead to the decrease of SS removal rate. Considering synthetically, the appropriate dosage of PAM was 20 mg/L.

4. Orthogonal experiment

There are many and complex factors affecting the flocculation effect of leachate\textsuperscript{[9,10]}. The PFSS and PAM dosage and pH value were selected as orthogonal experimental factors, excepting the interaction between the factors. According to the results of a single factor experiment, each factor was set at three levels, and the influencing factors and levels of the experiment are shown in table 1.

| Number | PFSS dosage (ml/L) /A | pH /B | PAM dosage (mg/L) /C |
|--------|-----------------------|-------|---------------------|
| 1      | 15                    | 6.5   | 15                  |
| 2      | 20                    | 7.0   | 20                  |
| 3      | 25                    | 7.5   | 25                  |

Aiming at the removal effect of COD, we designed an orthogonal experiment with 3 factors and 3 levels, according the condition of L9 (3\textsuperscript{3}) orthogonal table. 3 parallel experiments were performed under the same set of conditions, averaged for analysis. The results of orthogonal experiment and range analysis of leachate flocculation treatment are shown in table 2.
Table 2. Orthogonal test design and COD removal results.

| Number | A   | B   | C   | COD removal rate / (%) |
|--------|-----|-----|-----|-------------------------|
| 1      | 1   | 1   | 1   | 62.73                   |
| 2      | 1   | 2   | 2   | 59.88                   |
| 3      | 1   | 3   | 3   | 59.31                   |
| 4      | 2   | 2   | 3   | 63.93                   |
| 5      | 2   | 3   | 1   | 63.41                   |
| 6      | 2   | 1   | 2   | 70.72                   |
| 7      | 3   | 3   | 2   | 70.46                   |
| 8      | 3   | 1   | 3   | 75.43                   |
| 9      | 3   | 2   | 1   | 70.46                   |
| I      | 60.64 | 69.63 | 65.53 |
| II     | 66.02 | 64.76 | 67.02 |
| III    | 72.12 | 64.39 | 66.23 |
| R      | 11.39 | 3.38  | 1.49  |

According to the mean value range in the table 2, it can be seen that the order of factors influencing COD removal rate was PFSS dosage > pH > PAM dosage, indicating that the main influencing factors of flocculation settlement of landfill leachate were PFSS dosage and pH value.

In addition, it found through Analysis of Variance that the P values of PFSS dosage, pH value and PAM dosage were 0.002, 0.024 and 0.018 respectively in the range of test level, which were all less than 0.05, showing that the COD removal efficiency was significantly affected by the numbered A, B and C factor, and the results of range analysis were representative.

According to the results of orthogonal experiment and single factor experiment, the optimum parameters were as follows: the dosage of PFSS was 20 ml/L, pH 6.5, and the dosage of PAM was 20 mg/L. Set up three groups of parallel experiments to test the best parameters, and the results showed: When the concentration of COD, TP and SS in leachate was 1442 mg/L, 7.94 mg/L and 244 mg/L, the average COD concentration of effluent was 417 mg/L, and the average removal rate was 71.09%. The average concentration of effluent TP was 0.07 mg/L, and the average removal rate was 99.13%. The average concentration of effluent SS was 5 mg/L, and the average removal rate was 97.54%.

5. Conclusion

(1) Orthogonal experimental results of pretreatment of landfill leachate by polysilicate ferric sulfate flocculation process showed that the PFSS dosage had the greatest influence on the effluent effect of leachate flocculation treatment, and the effects of pH and PAM dosage decreased in turn. When the concentration of COD, TP and SS in landfill leachate is 1442 mg/L, 7.94 mg/L and 244 mg/L, respectively, the optimum parameters of flocculation process were as follows: PFSS dosage 20 ml/L, pH value 6.5, PAM dosage 20 mg/L, the removal rates of COD, TP and SS in effluent reached 99.13% and 97.54% respectively.

(2) It is necessary to add enough PFSS. Adequate H ion is beneficial to the Rapid release of CO₂ and mud-water separation of flocs, so as to avoid the influence of bubbles on the separation of mud and water in the late stage of flocs.

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Reference

[1] Huang Q R, Huo H H 2010 Application and respect of Magnetic flocculation and Magnetic separation technology J. Water supply and drainage 36 7 pp 150-152.
[2] Qiu Y Q, Sun S Y, Xiao X 2017 Deep removal of acidic complex Nickel by Magnetic flocculation coupled heavy Metal Catcher EDTC J. Chinese Environmental Science 37 2 pp 560-569.

[3] Wang C Z, Ren X F, Mei R W 2016 Application of loading Coagulation clarifier in double membrane reuse of dyeing Wastewater J. Water treatment Technology 42 2 pp 129-132.

[4] Liu D, Wang P, Wei D 2013 Removal of algal blooms from freshwater by the coagulation-magnetic separation method J. Environmental Science & Pollution Research International 20 1: pp 60-65.

[5] Zeng H F, Sun C B, W R 2011 Study on Magnetic flocculation Pretreatment of Landfill leachate J. Journal of Environmental Engineering 5 10 pp 2303-2306.

[6] Gao B Y, Song Y H, Yue Q Y 1997 Study on the Properties of Polysilicate Ferric Sulfate coagulant J. Environmental Science 18 2 pp 48-50.

[7] Ma Q S, Jia S, Sun L M 1988 Flocculation chemistry and flocculant (Beijing: China Environment Press).

[8] Zheng X H, Chen S Y, Zhong C Y 2004 Study on treatment of Landfill leachate by Polysilicate Ferric Sulfate J. Chemical Chronicle 18 11 pp 47-49.

[9] Yang C H, Song J Q 2001 The effect of ultrafine talcum powder on coagulation treatment of recycled paper and paper wastewater was studied by orthogonal test method J. Industrial Water treatment 21 7 pp 20-23.

[10] Cheng G W 2010 Water pollution control project (Beijing: Chemical Industry Press) pp 248-250.