Flocculation - Air Flotation Treatment of Wastewater from Paper-making Reconstituted Tobacco Sheet

Pengfei Li1,2,*, Qiangzhi Wu1,2, Hude Wu1,2, Shuangshuang Wen1,2, liang Wang1,2, Shuan Li1,2
1China Tobacco Hubei Industrial Co, Ltd, Wuhan 430000, China
2Hubei Xinye Reconstituted Tobacco Development Co, Ltd, Wuhan 430000, China

*Corresponding author e-mail: 370906347@qq.com

Abstract. In order to study the remove rate of organic pollutants in wastewater from paper-making reconstituted tobacco, and flocculation-flotation method was used to pretreat wastewater was discussed in this paper. The cationic polyacrylamide (CPAM) was used as coagulant, polymeric sulfate (PFS) and polyaluminium chloride (PAC) were used as flocculant. The effects of wastewater temperature, pH value, the type and dosage of flocculant and coagulant, the pressure and inflow of the air flotation on the flocculation and sedimentation of wastewater were analyzed. Research indicates: PFS was used with CPAM with relative molecular weight of 11 million and 40% ion degree, which had the best flocculation result. At the temperature of 32 ℃, the pH of 6.5, the PFS dosage of 400mg/L, the CPAM dosage of 5mg/L, the air flotation inflow of 150m³/h, the air flotation pressure of 0.35Mpa, the CODcr, SS and chroma removal rate are 65.2%, 87.43% and 72.53% respectively, which achieved the optimum effect. Under the optimum conditions, the operating cost of wastewater pretreatment by the method of flocculation – flotation was 2.72 yuan per ton.

1. Introduction
In the production of reconstituted tobacco sheets, 60-80 tons of waste water are usually discharged per ton of product [1-2]. The composition of waste water is complicated, which contains lignin, cellulose, nicotine, pectin, polysaccharides, and insoluble proteins. And many organisms that are difficult to be degraded by microorganisms [3-4]. The wastewater has a higher content of suspended solids, the loading of BOD and COD are heavier, darker shades (red-brown), higher redox potential, and perishability. Therefore, the efficient treatment of such wastewater is a problem that needs to be solved urgently in the development of reconstituted tobacco industry [5-6].

Flocculation-air flotation is one of the widely used technologies in wastewater treatment, which is often used as a wastewater treatment pretreatment unit [7-8]. The mechanism is to use the principle of high-efficiency shallow air flotation equipment to dissolve dissolved air into waste water. The small bubbles released in the dissolved air are used to float the suspended solids out of the water, and then the flocculant is added. The effect of flocculants such as electric neutralization, bridging, net capture, roll sweeping, and adsorption combined to disperse and flocculate the disperse phase in the wastewater to achieve SS removal [9-10].
In this experiment, the PFS and PAC were used as flocculants, and the three different types of CPAM were used as coagulants. The research study on the effect of flocculant, coagulant aid, pH, temperature, flow and air float pressure on waste water to obtained the optimum flocculation system and process parameters, which provides the technical support for the pretreatment of papermaking reconstituted tobacco wastewater.

2. Materials and Methods

2.1. Materials and instruments

(1) Raw material. The waste water was taken from the initial sedimentation pool of a tobacco sheet production enterprise in Hubei Province. The plant produces 1600-1800 cubic meters of wastewater per day. The primary and secondary wastewater treatment processes are shown in Figure 1. The wastewater quality indicators are shown in the table 1.

Flocculant

![Flowchart of reprocessing tobacco leaf wastewater](image)

| Section       | PH   | Temp(℃) | CODcr (mg/L) | BOD5 (mg/L) | SS (mg/L) | TN (mg/L) | TP (mg/L) | Chroma |
|---------------|------|----------|--------------|-------------|-----------|-----------|-----------|--------|
| Into water    | 6.8  | 30-35    | 11000-12000  | 5000-6000   | 4000-5000 | 142.8     | 92.5      | 2560   |
| Primary water | 6.8  | 30-35    | 8000-9000    | 3500-4000   | 2000-2500 | 138.5     | 89.6      | 2320   |

(2) Reagent. PFS is an industrial product, light yellow solid powder, total iron content ≥20%; PAM is an industrial product, white solid particles, and the three PAM properties selected are shown in Table 2. Potassium dichromate, silver sulphate, mercury sulphate, sulphuric acid, and sodium oxide are all analytically pure.

| Species | Types | Molecular weight | Ionization degree/% |
|---------|-------|------------------|---------------------|
| PAM1    |       | 450              | 15                  |
| PAM2    | cation| 800              | 30                  |
| PAM3    |       | 1100             | 40                  |

(3) Instrument. pH meter; Analytical balance; Electro thermal thermostatic blast drying oven; 722G visible light spectrophotometer; SHP-250 biochemical incubator; Plug colorimetric tube; ZR4-6 type six coagulator; SS-1Z suspension measurement; SS-1Z suspension analyser; 752 UV spectrophotometer.

2.2. Method

(1) Study the Effect of Flocculant on the flocculation of Wastewater
Take 200mL reconstituted tobacco wastewater from the initial sediment pool, the water PH is 6.8, the temperature is 30℃, stirring 2min(120r/min), then add different dosage flocculant PFS and PAC, continue stirring for 1 min at original speed, and then stir for 20min (30r/min) slowly, finally stand for 30min. Take the supernatant, determine the CODcr, SS, color removal rate.

(2) Study the Effect of Coagulants on the flocculation of Wastewater
Take 200mL wastewater from the initial sediment pool, stirring 2 min (120r/min), then add different amounts of three coagulants CPAM (Table 2). According to the results of above experiments, adding the optimal flocculant.

(3) Study the Effect of Temperature and PH on the flocculation of Wastewater
Take 200mL wastewater from the initial sediment pool, and adjusted the pH of water sample with NaOH or H2SO4, use electric furnace to control water sample temperature. According to the results of the above experiments, adding the optimal flocculant and Accelerator.

(4) Study the Effect of Inflow and Air Flotation Pressure on the flocculation of Wastewater
According to the results of the above experiments, adjust the PH and temperature of the water sample, adding the optimal type and amount of flocculant and Accelerator. Adjust the pump frequency and aeration to control the flow and the pressure.

3. Results and discussion

3.1. The Effect of PFS and PAC on the flocculation of Wastewater

![Figure 1-1](image1.png) Effect of SS removal rate on

![Figure 1-2](image2.png) Effect of COD removal rate on

![Figure 1-3](image3.png) Effect of chroma removal rate on
As can be seen from figure 1-1 and 1-2, with the increase of PFS and PAC, the compression of the double electric layer and the chances that particles take part in bridging and winding are increased, which is conducive to destroy the stability of waste water suspension colloid and then improve the removal rate of SS and COD. When the dosage of PFS reaches 400mg/L, the removal rate of SS and COD achieves a maximum value of 81.25% and 38.17%. When PAC is 400mg/L, the removal rate of SS reaches a maximum value of 73.45%. Beyond that, the COD removal rate reaches a maximum value of 31.36% when the PAC dosage is 450mg/L. However, when the flocculant continues increasing, the removal rate of COD is basically unchanged, even decreases. This is because when the amount of flocculant exceeds the isoelectric point of the colloidal destabilization, although the number of complex ions increases, the decrease of particle surface activity points which makes the adsorption bridge become more difficult. It may even cause turbidity and reduce coagulation effect [11].

Figure 1-3 shows that chroma removal rate reaches the maximum value when the PAC and PFS is 400 mg/L. To further increase dosage, the chroma removal rate of waste water drops rapidly with utilizing PFS. The main reason is that the PFS solution itself has obvious chroma, which has a very significant impact on the decoloration. In conclusion, when PFS is selected as the pretreatment flocculant for the reconstituted tobacco waste water, the suitable dosage is 400mg/L.

3.2. The Effect of the CPAM on the flocculation of Wastewater

![Figure 2-1 Effect of SS removal rate on](image)

![Figure 2-2 Effect of COD removal rate on](image)

![Figure 2-3 Effect of chroma removal rate on](image)
From figures 2-1~2-3 and 1-1~1-3, it can be known that the removal rate of SS, COD and chroma increase in different degree when three coagulant aids are added. The main reason is that the CPAM molecular chain has charge, which can compress the double electric layers on micro particle surface and improve the effect of flocculant adsorption and bridging [12-13]. Among them, the removal rates of SS, COD and chroma achieve the largest growth as 91.04%, 60.56%, 70.52% when CPAM2 is 5mg/L. While continuing increasing CPAM2, the removal rates of SS, COD and chroma basically remain unchanged, even fall. The main reason is that with the increment of CPAM, the COOH groups increase, which results in the increase of repulsion force between polymer chains and negatively electric particles in water and hinders the flocculation process [14]. Therefore, the compound use of the PFS and CPAM3 with a relative molecular weight of 11million and an ion degree of 40% could obtain the best flocculation effect.

3.3. The Effect of Temperature and pH on the flocculation of Wastewater

![Figure 3-1 Effect of PH on the flocculation](image1)

![Figure 3-2 Effect of Temperature on the flocculation](image2)

It can be seen from Figure 3-1, with the increase of pH value, the removal rate of wastewater SS, COD, and chroma increases first and then decreases. When pH = 6.5, the removal rates of SS, COD, and chroma were 91.18%, 63.43%, and 70.25%, respectively. Which the removal rate reaches a maximum. The reason for this phenomenon is because when the pH ≤4, Iron's high-priced multinuclear complex ion is converted to free iron ion and lose it agglomeration; As the pH becomes alkaline, The charge of aluminium ion hydrolysate is reduced and converted to [Al(OH)4]- anions. The charge repulsion will reduce the adsorption of suspended solids and destabilize the system [15].When the pH is 4.5-6.5, the chroma removal rate of the wastewater is stable. When the pH exceeds 6.5, the chroma removal rate drops sharply because the wastewater turns from reddish brown to dark black under strong alkaline conditions.

Figure3-2 shows that when the temperature is 32℃, the flocculation and sedimentation effect of wastewater is better, when the temperature continues to rise, SS, COD, chroma removal rate hasn’t changed much, the temperature is too high is also not conducive to the process of biochemical process.
3.4. The Effect of Inflow and Air Flotation Pressure on the flocculation of Wastewater

![Figure 4-1 Effect of inflow on the flocculation](image1)

![Figure 4-2 Effect of air pressure on the flocculation](image2)

As shown in Figures 4-1 and 4-2, According to the amount of sewage and the capacity of shallow air flotation, as the water inflow of shallow air flotation gradually decreases, the wastewater SS, COD, color removal rate increases gradually, and finally tends to stabilize. With the increase of air float pressure, the removal rate increased first and then decreased\(^{(16)}\). When the inflow was 150m\(^3/d\) and the air flotation pressure was 0.35Mpa, the removal rates of SS, COD and color reached the maximum, which were 65.2\%, 87.43\%, and 72.53\%, respectively. The reason for this phenomenon is that with the increase of the aeration volume of the air compressor, increased pressure of air flotation, the release amount of air bubbles in the dissolved air increases, the floatation of the suspended solids increases, and the removal rate increases. When the air float pressure exceeds 0.35 Mpa, the objects of flocculation in flotation tank will be agitated, reducing the removal rate and wasting power consumption.

3.5. The Analysis of operational Cost

Based on the above experimental results, the optimum flocculation system and optimized process parameters are: pH = 6.5, temperature is 32°C; the type of flocculant is PFS, dosage is 400mg/L; the type of coagulant is CPAM2, dosage is 5mg/L; the inflow of air floatation is 150m\(^3/h\), air float pressure 0.35Mpa. Taken together, the temperature doesn't need to be adjusted, and the pH can be adjusted by the hydrochloric acid.

Therefore, the operating cost of the flocculation-air flotation method for the pretreatment of reconstituted tobacco Wastewater includes the pharmaceutical charges and equipment power consumption. The pharmaceutical expenses include PFS, CPAM2, and consumption of HCl; the power consumption of equipment includes air-dissolving pump of high-efficiency shallow air flotation, slag, water distribution, scraper motor, and power consumption of various types of transfer pumps. Under the optimal conditions, the operating cost of the flocculant-flotation wastewater treatment agent is shown in Table 3.

| Material | Unit price (yuan/kg) | Dosage (kg/ton of water) | The cost for one ton of water (yuan) |
|----------|----------------------|--------------------------|-------------------------------------|
| PFS      | 1.70                 | 0.941                    | 1.599                               |
| CPAM2    | 23.73                | 0.0294                   | 0.698                               |
| HCl      | 0.47                 | 0.5                      | 0.235                               |
| Operating costs | -                 | -                        | 2.432                               |
In summary, under the optimal conditions, the operating cost of pretreatment of wastewater by flocculation-air floatation is 2.72 yuan/ton of water.

4. Conclusion
In this experiment, the flocculation-air floatation method was used to pretreat the waste water of reconstituted tobacco. The CPAM was used as a coagulant, PFS and PAC were used as flocculants. It studied on the effect of the temperature, pH, coagulants, flocculants, and inflow and air flotation pressure on the flocculation of waste water. The results show:

PFS is more suitable as a flocculant for reconstituted tobacco wastewater because of it has a higher removal rate of SS and COD.

The combination of CPAM and PFS with a relative molecular weight of 11 million and an ion degree of 40% has the best flocculation effect, and the removal rate of SS, COD and color of the reconstituted tobacco leaf waste is higher.

When the waste water temperature is 32°C, pH is 6.5; PFS dosage is 400mg/L; CPAM dosage is 5mg/L; air flotation equipment influent water 150m³/h, air float pressure 0.35Mpa, CODcr, SS, color The highest removal rates were 65.2%, 87.43% and 72.53% respectively.

Under the above-mentioned optimal conditions, the operating cost of pretreatment of wastewater by flocculation-air floatation is 2.72 yuan/ton of water.

References
[1] Chen Z G, Cai B, Wang J X. Comparison between domestic and foreign paper-process tobacco sheets [J]. Tobacco Science & Technology, 2002 (2): 4 - 10.
[2] Cao E H, Li J H. Research Progress in the Paper-making Process Reconstituted Tobacco Effluent Treatment and Reuse Technologies
[3] Chen R. Treatment on Wastewater from Production of Manmade Tobacco Leaf with Papermaking Method [J]. Environmental Science Survey, 2010.
[4] Hou Y, Li Y M, Zheng Z S. Analysis of pollutants in wastewater from manufacture of papermaking-reconstituted tobacco slice [J]. Journal of South China University of Technology, 2008, 36 (3): 95 - 98.
[5] Chen S Y, Li R L. Ozone Oxidation Pretreatment of Wastewater from Paper-making Process Reconstituted Tobacco Production [J]. Tobacco Science and Technology, 2014.
[6] Chen Z C, Jun L R, Wen Chao, et al. Study on Treatment of Reconstituted Tobacco Paper Wastewater [J]. Yunnan Chemical Technology, 2014, 16: 15 - 18.
[7] Wang H T, Liu H B, Cui Z Q, et al. Study on Deep Treatment of Paper Making Waste Water by Using Flocculation Precipitation Process [J]. Pollution Control Technology, 2012, 1 (2): 23 - 27.
[8] Pinto Filho A C, Brandao C C. Evaluation of flocculation and dissolved air flotation as an advanced wastewater treatment. [J]. Water Science & Technology A Journal of the International Association on Water Pollution Research, 2001, 43 (8): 83 - 87.
[9] Tian C N, Jie L I, Wang Y E, et al. Application of shallow air flotation for water treatment[J]. Industrial Water & Wastewater, 2013, 2 (1): 17 - 20.
[10] Kwon H, Jung C, Lee J. Dissolved air flotation (DAF) method for marine microalgae using chemical flocculants [J]. 2012.
[11] Luo L. Study on the Wastewater Treatment of the Paper Production by PAF and PAM Coagulant [J]. Anhui Chemical Industry, 2005.
[12] Razali M A A, Ahmad Z. Treatment of pulp and paper mill wastewater with various molecular weight of poly induced flocculation [J]. Chemical Engineering Journal, 2011, 166 (2): 529 - 535.
[13] R Kothari, VV Pathak, APandey. A novel method to harvest Chlorella sp. via low cost bioflocculant: Influence of temperature with kinetic and thermodynamic functions [J]. Renewable Energy Resources, 2017, 225: 84 - 89.
[14] Tang H J. Inorganic polymer flocculation theory and flocculant [M]. Beijing: China Building Industry Press 2010.

[15] Liang Z, Wang Y, Zhou Y, et al. Variables affecting melanoidins removal from molasses wastewater by coagulation /flocculation [J]. Sep. Purif. Technol, 2009, 68 (3): 382 - 389.

[16] J A Filho, A Azevedo ,R Etchepare ,J Rubio. Removal of sulphate ions by dissolved air flotation (DAF) following precipitation and flocculation [J]. International Journal of Mineral Processing, 2016, 149: 1 - 8.