Study on pre-treatment of pita wastewater with sic membrane

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Abstract. Under the conditions of temperature of 20°C and transmembrane pressure of 0.2MPa, a silicon carbide membrane with a pore size of 0.04μm was introduced to treat pure terephthalic acid (PTA) wastewater with the dead end filtration method. The membrane was found to exhibit high flux as 1987L/(m²·h), the solid content decreased from 0.501% to 0.003%, the COD decreased from 23345.40mg/L to 17862.11mg/L. The flux recovery rate was achieved 97.6% and 100% after back flushing and membrane cleaning respectively.

1. Introduction

Pure terephthalic acid (PTA) is produced by high temperature oxidation of p-xylene to produce crude terephthalic acid, followed by hydrotreating. In the refining stage of PTA, a large amount of deionized water is needed as a solvent, and most of the deionized water leaves the main process flow as a mother liquor and becomes waste water, and carries a large amount of suspended solids (composition is PTA) whose particle diameter distribution is 700-2000 nm.

At present, PTA wastewater treatment methods mainly include chemical precipitation method [1-5], chemical oxidation method [6-9], adsorption separation method [10-12], biochemical oxidation method [13-19] and filtration separation method [20-21] etc. Chemical precipitation treatment of wastewater requires the introduction of new chemical substances, which can easily cause secondary pollution. The chemical oxidation method has strict application conditions and high processing costs. Adsorption separation requires a low solids content of the wastewater, and regeneration of the adsorbent makes the process relatively complicated. The biochemical oxidation process has a long cycle time, high material consumption and energy consumption. Filtration separation treatment of the wastewater can effectively recover the solid particles in the wastewater, reduce the COD content, but the low pH, high temperature and other characteristics of the wastewater limits the use of most membranes. Silicon carbide membrane has the advantages of high temperature resistance, acid and alkali resistance, narrow pore size distribution range, large flux, thus is suitable for treating PTA wastewater.

In this work, silicon carbide membrane is used to pre-treat PTA wastewater. The membrane-treated permeate is further processed and returned to the refining unit. The concentrate returns to the pressure filtration process to recover PTA solids. The results showed that, most of the solid particles were trapped and the flux recovered after membrane cleaning.
2. Experimental

![Schematic Diagram of Membrane Separation](image)

**Figure 1.** The schematic diagram of the membrane separation

A cross-flow membrane separation system (DJ-SJ-02, Hubei Dixie Membrane Technology Co., Ltd.) was employed as illustrated in Figure 1.

Gravimetric method (National Standard GB 11901-89) is used to analyze the Determination of suspended solids in raw material and permeate. Water quality-Determination of the chemical oxygen demand-Dichromate are analysed with Potassium Dichromate Method (HJ 828-2017). Particle diameter of solid is tested with Malvern Laser Particle Size Analyser (Malvern MASTERSIZER 3000).

The cleaning method for the experimental membrane was as follows: 20L of water was added, and 100g of surfactant was added thereto for a total of 60 minutes. The transmembrane pressure was 0.22MPa in the first 30 minutes. Then the effluent valve was opened. The transmembrane pressure was 0.2 MPa. After the cleaning was completed, clear water was added.

3. Results and Discussion

3.1. Particle diameter of solid

The selection of pore size of the membrane is determined by the diameter of the solid particles. If the pore size is too smaller, the solid particles will clog the pores of the membrane, causing a drop in flux. If the membrane pores are too larger, solid particles will pass through the membrane pores and will not achieve filtration. As shown in Fig 2 below, the diameter of the solid particles in the wastewater was distributed in 2~13μm, the smallest particle diameter was 2.47μm, the largest particle diameter was 12.3μm, and the median size was 7.09μm. In order to achieve a good filtration effect and efficiency, we chose a silicon carbide membrane with an aperture of 0.04μm to pretreat PTA wastewater.

![Distribution of Particle Diameter](image)

**Figure 2.** Distribution of particle diameter
3.2. Effect of transmembrane pressure on membrane flux

Figure 3 shows the effect of transmembrane pressure difference on membrane flux at a temperature of 20°C. As can be seen from Fig. 3, when the transmembrane pressure difference raised from 0.1MPa to 0.3MPa, the membrane flux increased from 1304L/(m²·h) to 2530L/(m²·h). Considering the pressure resistance and sealing performance of the membrane module, we selected 0.2MPa for the transmembrane pressure.

![Figure 3. The schematic diagram of the membrane separation](image)

3.3. Filtering effect

The test results of COD and suspended solids content of the feed batches and the dialysate of the experimental batches are shown in Table 1 and Table 2. It can be seen that after Sic membrane treatment, the effluent solids content decreased from 0.501% to 0.003%, and the COD decreased from 23345.40mg/L to 17862.11mg/L. This shows that most of the solid particles were intercepted.

| Beaker (g) | Sample (g) | Beaker and sample (g) | Solid content (%) |
|------------|------------|----------------------|------------------|
| 1          | 17.1645    | 11.2369              | 17.2203          | 0.501             |
| 2          | 17.8963    | 13.2487              | 17.9623          | 0.498             |
| 3          | 19.8622    | 10.2621              | 19.8625          | 0.003             |
| 4          | 16.7121    | 13.8289              | 16.7124          | 0.002             |

Table 2. The quality of feed water and the permeate

| Temperature (°C) | Solid Content (%) | COD (mg/L) | Removal rate Solid intent |
|------------------|-------------------|------------|---------------------------|
| 20               | 0.501             | 0.003      | 23345.40                  | 17862.11          | 99.4%          |

3.4. Back flushing of ceramic membrane

During back flushing, the module was backwashed by the clean liquid returning with compressed air, thereby forcing the particles on the membrane surface and pores back into the retentive and destroying the gel layer on the membrane surface and the concentrated polarization layer, thereby increasing the flux. After back flushing (3 seconds per 30min, under 0.3Mpa), the membrane flux changed as shown in Fig 4 and the membrane flux recovery rate was 97.6%.
3.5. Ceramic membrane cleaning
Membrane fouling, which results from specific interactions between membrane pore surface and solid particle, caused a rapid flux decline of the whole membrane. In order to reduce the effect of membrane fouling and to recover membrane flux, a periodic hydraulic backwash process was employed in our study. Fig. 4 shows the effect of backwashing on permeation flux performance. Higher flux restoration efficiency was observed for the Sic membrane in each run. It is realized to recover up to 1987 L/(m²·h), over 1947 of the original flux, 1940 L/(m²·h) at the second cycle, during this step, flowed backwards through the membrane pore-structure, resulting in an effective removal of solid particle that adhered on membrane surface. It also can be seen from Fig 5 that a periodic backwashing was effective to maintain its flux recovery performance, and 100% flux recovery efficiency of the original flux was observed.

4. Conclusion
The PTA wastewater was treated by dead-end filtration under the conditions of transmembrane pressure 0.2MPa and temperature 20°C for a full silicon carbide membrane with a pore size of 0.04μm. The results showed that the membrane flux could be achieved 1987 L/(m²·h), and the effluent solid content decreased from 0.501% to 0.003%. The COD decreased from 23345.40mg/L to 17862.11mg/L. The flux recovery rate was achieved 97.6% after back flushing, 100% after membrane cleaning.
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