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Preparation of polyethylene porous tube membrane and fouling mechanism during the filtration process

Zhangfu Yuan[a], Lu Mei[a], Shanshan Xie[a], Linfei Zhao[a], Hongxin Zhao[a,b,*], Bingsheng Xu[c]

[a]*Collaborative Innovation Center of Steel Technology, University of Science and Technology Beijing, Beijing 100083, China, Tel. +86 13718113630; email: hxzhao_ustb@163.com (H. Zhao), Tel. +86 18911786560; email: zfyuan2016@ustb.edu.cn (Z. Yuan), Tel. +86 15229208590; email: 15229208590@163.com (L. Mei), Tel. +86 13149497839; email: 729055633@qq.com (S. Xie), Tel. +86 18811082667; email: zhaolinfei2021@163.com (L. Zhao)*

[b]*National Engineering Research Center of Green Recycling for Strategic Metal Resources, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China*

[c]*Research Branch of Resource and Environment, China National Institute of Standardization, Beijing 100191, China, Tel. +86 13718113630; email:374486755@qq.com*

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ABSTRACT

Polyethylene porous filter tube membranes were prepared by powder sintering method. The microstructure of the tubular membrane, the effect of inorganic particles, and emulsified oil on membrane fouling were studied. The membrane flux variation curves were analyzed to further reveal the fluxing mechanisms. The results showed the membrane was composed of particles with the size of 10–60 μm, and there were many irregularly shaped three-dimensional through pores with the size of 20–70 μm in its cross-section. For inorganic particle dispersion filtration, the larger the dispersion concentration was, the greater the membrane flux dropped, and the smaller the irreversible fouling was; the smaller the inorganic particle size and the higher the filtration flow rate were, the greater the membrane flux dropped and the greater the irreversible fouling were; The analysis showed that at the beginning of filtration, the membrane first underwent complete pore blocking, and as the filtration process proceeds, the contamination pattern of the membrane changed, and cake filtration blocking occurred. For emulsified oil filtration, the larger the pH value was, the smaller the zeta potential, and the smaller the emulsified oil was, the greater the decreased in membrane flux and the greater the irreversible fouling was. For the filtration of turbid ring water with inorganic particles and emulsified oil, the membrane flux decreased less than the filtration with emulsified oil only; as the concentration of emulsified oil increased, the irreversible fouling tended to rise.

Keywords: Porous filtration membrane; Membrane fouling; Inorganic particles; Emulsified oil; Polyethylene

1. Introduction

Water resources are necessary for human development and an important part of living organisms. The current global water shortage and pollution problems are seriously affecting the sustainable development of human society and causing widespread concern all over the world. As a resource-based industry with high water consumption and pollution, the metallurgical industry uses a huge amount of new water and wastewater. Realizing the rational use of recycled industrial water and reducing external discharge is of great significance to realize the rational use of water resources and develop a green steel industry [1–3]. The turbid ring water produced by the continuous casting process of iron and steel has a high content of pollutants,

* Corresponding author.

containing many different particle sizes of iron oxide particles, metal dust, and grease [4]. These continuous casting turbid ring water, if not treated, is directly discharged or directly recycled, not only endangers aquatic resources and human health but also brings great harm to continuous casting production [5,6]. Therefore, whether the iron oxide, dust, and grease in the turbid ring water can be effectively separated is a prerequisite to deciding whether the turbid ring water can be recycled and whether the water resources can be reasonably utilized.

At present, the traditional treatment method of continuous casting turbid ring water is to use physical methods such as sedimentation tank and high-speed sand filter to reduce the suspended matter and oil in turbid ring water, but it has the disadvantages of unsatisfactory filtration effect, frequent replacement of filter media, no oil removal, and high operation cost, and is not easy to replace and maintain, which seriously affects the production of steel [2]. In recent years, with the development of polymeric materials and an emerging membrane treatment technology [7], membrane filters developed with polymeric materials as filter media have gradually replaced traditional filters and are increasingly used in industrial wastewater fields such as metallurgy, chemical industry, electroplating, printing, and dyeing [8,9].

Membrane separation technology uses selectively permeable membrane as the separation medium to achieve the separation, purification, and concentration of different components with the help of external energy or chemical potential difference [10,11], which has the advantages of high filtration precision, high separation efficiency, simple operation, low energy consumption, no secondary pollution, simple backwashing, and low operating cost, etc. It is considered as one of the most promising and promising technologies in the 21st century [12,13]. However, in the process of treating turbid ring water with membrane technology, iron oxide particles, metal dust, and grease in turbid ring water have physicochemical and mechanical interactions with the membrane, which are easily absorbed and deposited on the membrane surface and inside the membrane pores, causing the membrane pores to be clogged or smaller, resulting in membrane fouling, which decreases the membrane flux, filtration efficiency, and precision, and also shortens the membrane service life and increases the treatment cost [14,15]. Therefore, it is necessary to research the mechanism of the membrane fouling to promote the development of the membrane separation technology and the overall direction of continuous performance improvement.

In this research, polyethylene porous filter tube membrane was prepared by powder sintering method using ultra-high molecular weight polyethylene as raw material to research its microstructure, anti-fouling performance, etc. were studied. The actual purification process of turbid ring water of continuous casting was simulated by experiment. The effects of concentration, particle size, and filtration flow rate of inorganic particles dispersion system on membrane fouling were studied; the effects of pH and size of emulsified oil on membrane fouling were studied; the microstructure, filtration performance, and fouling characteristics of the membrane during the filtration of turbid ring water mixed with inorganic particles and emulsified oil were analyzed in depth. The membrane flux variation curves were analyzed to further reveal the contamination characteristics and mechanism of polyethylene porous tube membranes.

2. Experimental

2.1. Materials

The materials and reagents required for the experiments are shown in Table 1.

2.2. Preparation process

2.2.1. Preparation of polyethylene porous tube membrane

Polymer polyethylene, aluminum stearate, hydroxypropyl methylcellulose, and rare earth oxides are added to

Table 1
Main materials and reagents for the experiments

Material	Specification	Manufacturers
Ultra-high molecular weight polyethylene	<100 mesh	Macklin
SiO_2	600–800 mesh	Shanghai Macklin Biochemical Co., Ltd
Fe_3O_4	70 μm	Nangong Jingrui Alloy Products Co.
Fe_3O_4	55 μm	Nangong Jingrui Alloy Products Co.
Fe_3O_4	35 μm	Nangong Jingrui Alloy Products Co.
Fe_3O_4	5 μm	Nangong Jingrui Alloy Products Co.
Anhydrous ethanol	Analysis pure	Sinopharm Group
CTAB	Analysis pure	Aladdin
SDS	Analysis pure	Shanghai Macklin Biochemical Co., Ltd
Tween-80	Analysis pure	Aladdin
Aluminum tristearate	–	Shaoyang Paradise Auxiliary Chemical Co.
Hypromellose	–	Shandong Yiteng New Material Co.
Deionized water	–	–
Gear oil	Industrial grade	–

a ball mill and grounded for 5–6 h. The grounded mixture is added to a tubular stainless-steel mold, and after oscillating and mixing, the mold is placed in a sintering chamber and sintered at 220°C for 2–3 h. Finally, the polyethylene porous filter tube is obtained by cooling in the furnace to room temperature.

2.2.2. Preparation of inorganic particle dispersion system and emulsified oil

Preparation steps of inorganic particles dispersion system: different particle sizes and weights of Fe_3O_4 and SiO_2 are weighed, then they are added into 250 L of deionized water and stirred at high speed for 30 min with an electric speed control stirrer to obtain the inorganic particles suspension dispersion system.

Preparation steps of emulsified oil: a certain amount of GL-5 gear oil and surfactant CTAB, SDS, and Tween 80 (the mass ratio of the two is 10:1) are weighed, then added to 250 L of deionized water. Finally, electric speed stirrer is used with high-speed stirring for 60 min to produce a milky white emulsified oil.

2.3. Experimental method

The flow chart of the experimental setup used for the polyethylene porous tube membrane filtration system is shown in Fig. 1, where the polyethylene tube membrane is directly submerged in the Plexiglas vessel. Raw water enters the Plexiglas vessel through a screw pump and flows out from the outer side of the membrane tube into the inner side. The inlet and outlet are equipped with flow meters and pressure gauges, respectively, and the outlet is equipped with a backwash pump. The bottom of the Plexiglas vessel is equipped with a drainage port, and the membrane contaminants after backwashing are discharged from the drainage port. In this experiment, constant pressure dead-end filtration is used, and the filtrate volume is recorded every 60 s. During the filtration experiments, the pressure of the filtration system is regulated using an air compressor and a pressure reducing valve, and the pressure of the filtration system is monitored in real-time by a pressure gauge. The pressure used for the experiment is 0.3 MPa. After 2,400 s, filtration is stopped, backwashing is carried out, the backwashing time is 5 min, and the next filtration cycle is carried out after flushing. During the backwash test, the filtration system is stopped and the water obtained from the previous experimental filtration flows into the filter in the direction of the black dashed arrow in Fig. 1 for backwashing the membrane by means of a backwash pump and an air compressor, and the backwash pressure and the filtration pressure are kept the same.

This experiment uses the membrane flux decline and reversible and irreversible membrane fouling of polyethylene porous tube membrane to characterize the polyethylene porous filter tube membrane. The membrane flux refers to the amount of fluid passing through the unit area per unit time, usually expressed as J. The filtration area of the membrane during the filtration experiment is 0.387 m^2. The membrane fouling formed during the membrane filtration process is divided into reversible membrane fouling (RF) and irreversible membrane fouling (IF). Reversible membrane fouling can be removed by backwashing to achieve recovery of the membrane flux. Irreversible membrane fouling cannot be cleaned off by hydraulic backwashing; the sum of reversible and irreversible membrane fouling is called total membrane fouling (TF). According to Darcy's law, its calculation formulas are as follows.

$$J = \frac{V}{At} \tag{1}$$

where J denotes filtration flux (m/s), V denotes filtration volume (m^3), A denotes effective filtration membrane area (m^2), and t denotes filtration time (s).

$$\text{RF}_n = \frac{J_{s(n+1)} - J_{e(n)}}{J_n};\ \text{IF}_n = \frac{J_n - J_{s(n+1)}}{J_n};\ \text{TF}_n = \text{RF}_n + \text{IF}_n \tag{2}$$

where RF_n, IF_n, and TF_n denote, the reversible membrane fouling, irreversible membrane fouling, and total

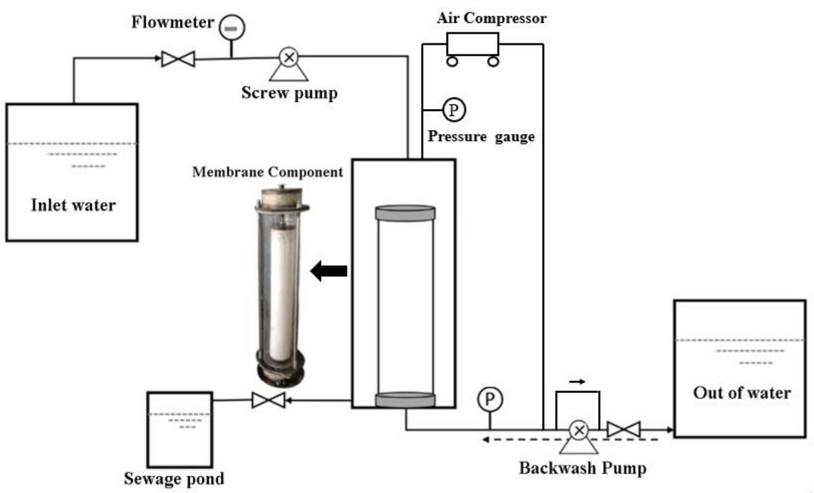

Fig. 1. Schematic diagram of submerged polyethylene porous membrane filtration system.

membrane fouling of the n cycle, respectively. J_n denotes the initial pure water flux of the n cycle of the polyethylene tubing membrane, $J_{e(n)}$ denotes the terminal membrane flux at the end of the n cycle of the polyethylene tubing membrane, and $J_{s(n+1)}$ denotes the initial pure water flux of the n+1 cycle of the polyethylene tubing membrane.

Retention rate is one of the important indicators for evaluating membrane filtration performance. The turbidity of permeate before and after filtration is measured by using turbidity meter. The retention rate of the membrane is calculated using the formula as follows [16]:

$$R = \left(1 - \frac{C_p}{C_0}\right) \times 100\% \quad (3)$$

where R is the retention rate (%), C_p is the turbidity of the permeate after filtration (NTU), and C_0 is the turbidity of the permeate before filtration (NTU).

2.4. Characterization methods

Field emission scanning electron microscope (FESEM) (JSM-7001F) was used to characterize the microscopic morphology of polyethylene porous tube membrane. The pore size and distribution of the membrane were determined using an automated mercury pressure method (Autopore IV 9500). The turbidity of the solutions before and after filtration was analyzed using a benchtop turbidity analyzer (NTU-2S000). Liquid pH was measured using a pH meter (6175-3C/FEP20). Oil droplet size and zeta potential values of emulsified oil were measured using a nanometer particle size and zeta potential analyzer (DelsaNano C).

3. Results and discussion

3.1. Microstructure of polyethylene pipe membrane

The size of the particles of the filtration material itself and the size and of the formed pore size have a crucial influence on the filtration performance such as filtration flux and filtration efficiency [17–19]. Fig. 2 shows the scanning electron microscope image of the microstructure of the outer wall of the polyethylene porous filter tube film, from which it can be seen that the polyethylene porous filter tube is formed by the accumulation of particulate matter with a particle size of about 10–60 μm, and there are a large number of irregularly shaped three-dimensional through pores with a pore size of about 20–70 μm. It can be seen from Fig. 2a and b that, due to the extrusion of the die during the production process, the polyethylene porous filter tube shows a flattened shape on the outer side of the spherical particles on the outer wall. During the filtration process, the impurity particles with particle size smaller than the membrane pore size will be adsorbed within the pores, causing irreversible fouling of the polyethylene porous filter membrane tube, and the impurity particles larger than the most available pore size will easily form a filter cake layer on the membrane surface.

3.2. Fouling characteristics of polyethylene porous tube membrane

Polyethylene porous filter tube membrane in the process of filtration of turbid ring water is accompanied by membrane flux decline, membrane fouling, flux recovery rate reduction, and other problems, which in turn affect the service life of the membrane. By analyzing the concentration, particle size, and filtration flow rate of inorganic particles dispersion system; the pH and size of emulsified oil; the influence of turbid ring water with inorganic particles and emulsified oil coexisting on membrane fouling, the fouling mechanism of polyethylene porous tube membrane is studied in depth to control membrane fouling effectively from the root.

3.2.1. Influence of inorganic particle dispersion system on membrane fouling situation

Continuous casting turbid ring water contains a large amount of iron oxide, metal dust, grease, and other impurities, where the concentration of suspended iron oxide can be as high as 1500 mg/L, with particle sizes ranging from 20 to 60 um [4]. Therefore, a detailed analysis of the membrane fouling by inorganic particles is required.

The total number of particles in the dispersion system determines the degree of fouling of polyethylene pipe membrane. With the same mass fraction of inorganic particles, the higher the concentration is, the higher the total number of particles are. The effect of inorganic particle concentration

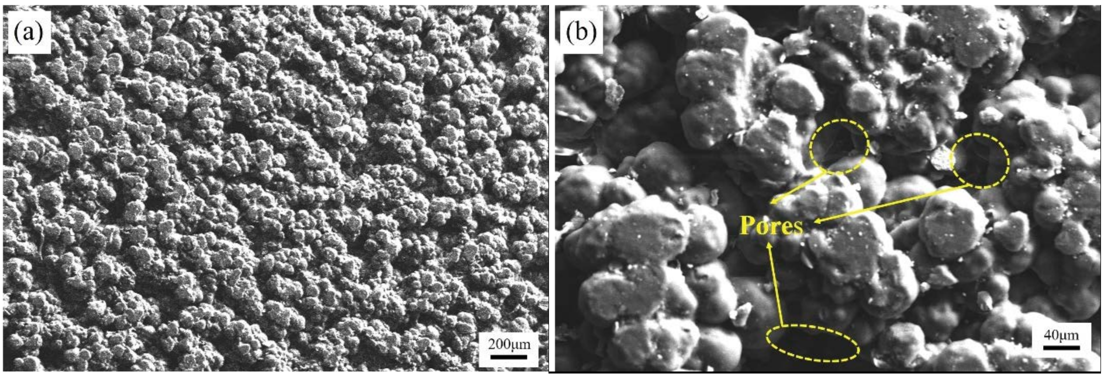

Fig. 2. Micro-SEM images of porous polyethylene tube membrane: (a) outer wall and (b) enlarged view of outer wall.

(0.15, 0.3, and 0.45 kg/m³) on the fouling of the membrane during the filtration process of the inorganic particle dispersion system is shown in Fig. 3. As the concentration of the dispersion system increases, the flux decreases more and more by 16.1%, 20.7%, and 26.8%, respectively, as shown in Fig. 3a. At the beginning of filtration, the flux decreases rapidly and gradually stabilizes when the flux decreases to a certain value, which is because, at the beginning of filtration, the smaller particles will be absorbed and deposited in the membrane pores, making the flux decrease rapidly. With the extension of filtration time, the particles will accumulate on the membrane surface layer by layer to form a filter cake layer, preventing the smaller particles from further entering the membrane pores, thus stabilizing the flux. From Fig. 3b it can be seen that as the concentration of the dispersion system increases, it causes the reversible fouling of the membrane to increase, the irreversible fouling to decrease, and the total membrane fouling to increase. The increase in total membrane fouling indicates that the more serious the membrane fouling is, the more the membrane flux decreases simultaneously. The reversible fouling can be removed by backwashing with water streams, thus restoring the membrane flux [20–22]. During filtration, a more concentrated dispersion system is more likely to form a cake layer on the membrane surface than a less concentrated dispersion system, resulting in larger reversible fouling that is easily removed by backwashing but will prevent smaller particles from entering the die pores and alleviate irreversible fouling. For the particulate matter dispersion system, the concentration of particulate matter in the water sample before and after filtration is an important index to evaluate the membrane filtration performance. As seen in Fig. 3c, the turbidity before filtration is 41, 71, and 88 NTU, and the turbidity after filtration is 2, 4, and 7 NTU for different concentrations of the dispersion system, and the retention rates are 95.1%, 94.4%, and 92%, respectively. With the increase of inorganic particle concentration, the retention rate decreases but still remains above 90%, indicating that polyethylene porous tube membrane has good filtration performance for particulate dispersion system.

The drop of membrane flux and fouling for different filtration flow rates for inorganic particulate dispersion system filtration are shown in Fig. 4. The flow rate of the dispersion system during filtration greatly influences the formation of the filter cake layer and the permeation behavior of the particulate matter [23], which affects the fouling behavior of the membrane. It can be seen from Fig. 4a that as the filtration flow rate increases, the flux decreases more, with the decrease rates of 7.8%, 21.5%, and 34.8%, respectively. The higher the filtration flow rate is, the mall

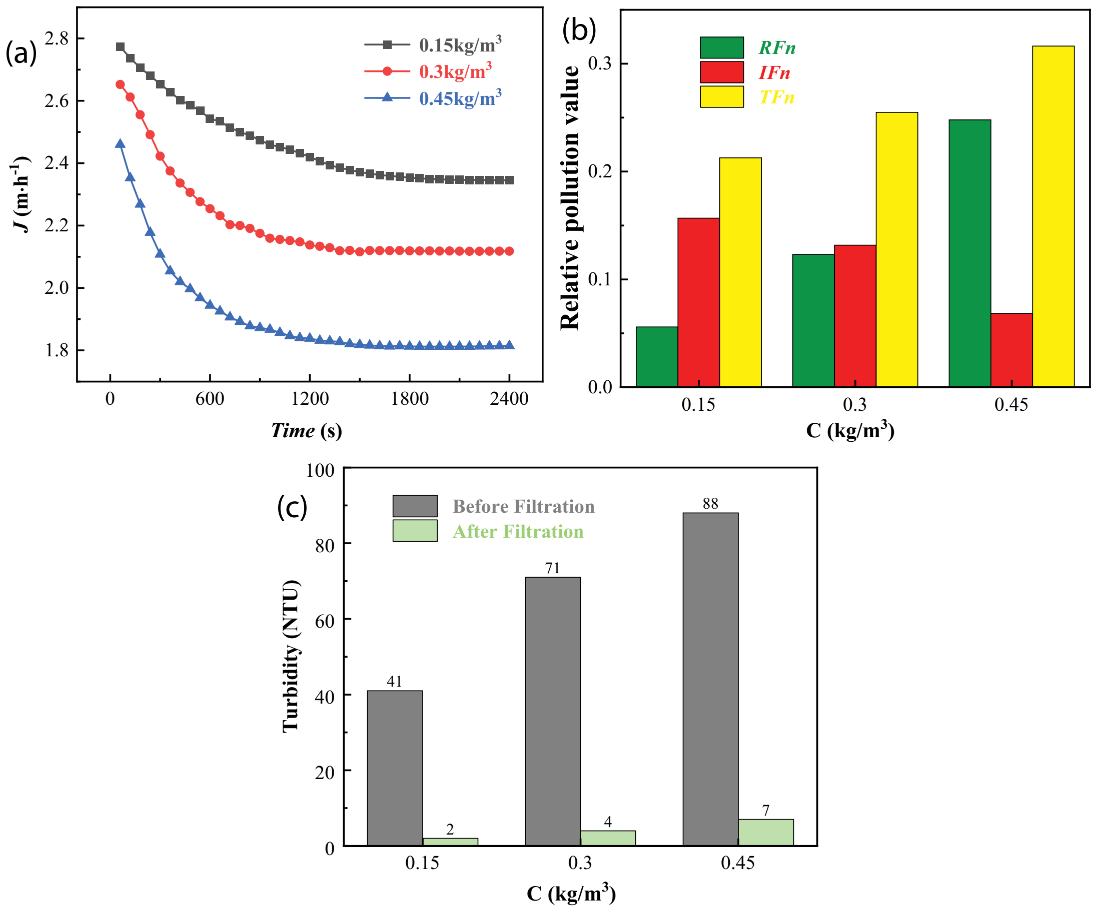

Fig. 3. Effect of different inorganic particle concentrations on the fouling behavior of the membrane: (a) curve of the membrane flux, (b) fouling of the membrane, and (c) change in turbidity before and after filtration.

particles enter the membrane pore at the early stage of filtration, and the faster the formation of the filter cake layer is. At the same time and concentration, the more the total number of particles accumulated on the membrane surface is, the more the cake layer formed on the membrane surface will be compressed by the water flow, and the more the membrane flux will decrease as the filtration time increases. Fig. 4b shows the fouling characteristics of the membrane under different filtration flow rates. With the increase of filtration flow rate, the more serious the reversible, irreversible, and total membrane fouling are. It is because the larger the flow rate is, the more impurity particles reach the membrane surface and pore size at the same time. The impurity particles are trapped by the porous filter tube membrane when passing through the membrane pores and surface so that they are deposited and adsorbed on the membrane surface, thus forming a filter cake layer. And the thicker cake layer in the filtration process has a serious impact on the membrane fouling, which will cause a larger flux drop and affect the filtration efficiency. It indicates that the lower flow rate in the filtration process has an important role in reducing membrane fouling.

Particle size has a significant effect on membrane fouling [24]. Fig. 5 shows the membrane flux variation and membrane fouling caused by inorganic particles of different particle sizes at a dispersion system concentration of 0.3 kg/m^3 and a flow rate of 0.4 m^3/h in the filtration process. The total number of particles in the dispersion system greatly influences the degree of fouling of the polyethylene tube membrane, and the smaller the particle size is, the more the total number of particles is at the same concentration. From the flux drop curve in Fig. 5a, it can be seen that the membrane flux decreases by 8%, 16%, 18.8%, and 26.5% as the particle size of inorganic matter decreases. It indicates that the membrane flux decreases more as the particle size of the dispersion system decreases during the filtration process, and the total fouling of the membrane increases as the number of particles increases. From the analysis of fouling reversibility of the membrane in Fig. 5b, it can be seen that as the particle

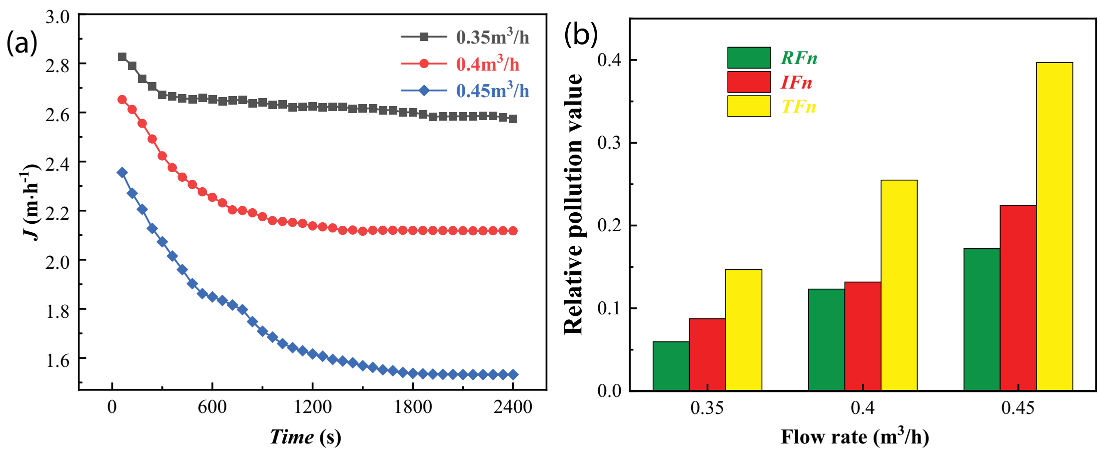

Fig. 4. Effect of different filtration fluxes on the fouling behavior of the membrane: (a) curve of the membrane fluxes and (b) fouling of the membrane.

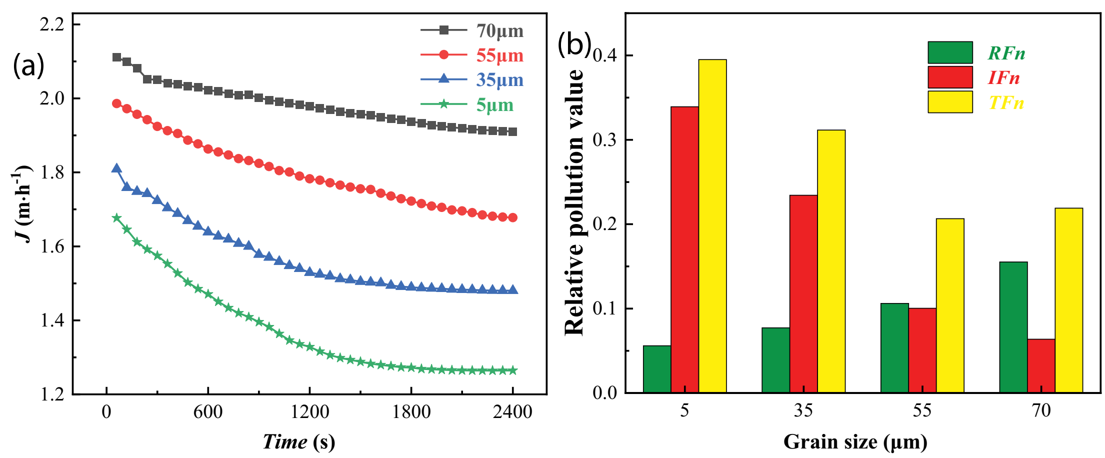

Fig. 5. Effect of different inorganic particle sizes on the fouling behavior of the membrane: (a) curve of the membrane fluxes and (b) fouling of the membrane.

size of the dispersion system increases, irreversible fouling decreases, and reversible fouling gradually increases. The reduction of irreversible fouling decrease is due to the dispersion system with small particle size forming a thin layer of filter cake, which is easily compressed by the water flow closely and is not easily removed by the backwashing process. In addition, the particles with small particle size will pass through the membrane surface and enter the membrane pores for adsorption and deposition during the filtration process, thus increasing the irreversible fouling. The gradual increase in reversible fouling is because fewer dispersion system particles of large size enter the membrane interior, and the filter cake formed on the membrane surface is loose and easily removed by backwashing. The reversible fouling caused by 70 μm is the most serious, and the irreversible fouling caused by the smallest particle size of 5 μm is the largest. The smaller the particle size and the greater the number of particles at the same concentration are, the greater the total membrane fouling cause. It indicates that the particle size of suspended particles in the wastewater affects membrane fouling.

During the filtration process for sewage, the fouling of the membrane is not a single fouling mode, and membrane fouling cannot be avoided. It impacts the stability of the water purification process, shortens the life of the tube membrane, and increases the energy consumption of the water purification process. The inorganic particles of the sewage significantly impact the fouling of the membrane. According to Wu's study [25–27], Figs. 3–5 were analyzed. During the filtration cycle, the membrane first occurs complete pore blocking in the early filtration stage. As the filtration process proceeds, the particulate matter will accumulate layer by layer on the membrane surface, and the membrane surface gradually forms a filter cake layer, the fouling mode of the membrane is transformed and begins to take place cake filtration. Overall, during the filtration process, complete clogging fouling occurs in the membrane first, followed by cake clogging fouling.

3.2.2. Effect of emulsified oil on the membrane fouling situation

pH is one of the important indexes to evaluate the wastewater system, and it has an important influence on the zeta potential of colloidal particles in emulsions [28,29]. Therefore, in this paper, the zeta potential and size of emulsified oil droplets at different pH values were examined by zeta potential meter, and the membrane flux variation law under different pH conditions was investigated.

The zeta potential and size of emulsified oil droplets at different pH values are shown in Fig. 6. It can be seen that the zeta potential of oil droplets gradually decreases as the pH values increases. When the pH value is 6, the zeta potential is the largest, about –47.5 mV. It is due to the fact that the oil droplet itself is negatively charged, and when the pH value is 6, the H^+ concentration increases, and some H^+ is adsorbed on the surface of the oil droplet, which makes the zeta potential increase [30]. When the pH value is 7, the oil droplet size is the largest, and there is a small decrease in the oil droplet size as the pH value increases. It is because the pH value increases, the zeta potential of oil droplets decreases, the electrostatic repulsive force between oil droplets increases, and the chance of oil droplet collision and polymerization decreases, so the oil droplet size is smaller.

The pH of the effluent system has an important influence on the membrane fouling behavior in the filtration process. Fig. 7 shows the change curve of the membrane flux of polyethylene porous filter tube membrane in the filtration of emulsified oil at different pH value. It can be seen from the figure that with the increase of pH value, the membrane flux decreases gradually, which indicates that the influence of pH on the membrane filtration process is more obvious. With the increase of pH value, the zeta potential and the size of oil droplets decrease gradually. The smaller oil droplets are easier to enter into the pores of porous filter tube for strong adsorption. The decrease in zeta potential causes the membrane to increase the repulsive force on the oil droplets, which makes an oil membrane form on the membrane

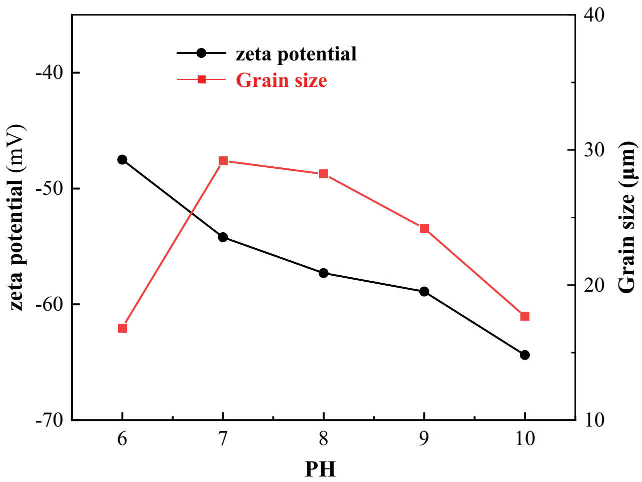

Fig. 6. Effect of pH on zeta potential and particle size of emulsified oil.

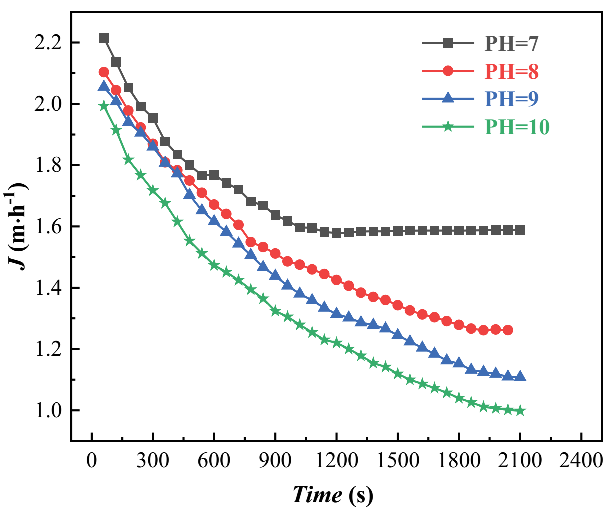

Fig. 7. Curve of the membrane flux under different pH conditions.

surface [31]. In the middle and late stages of filtration, the small-sized oil droplets cannot enter the membrane pores and accumulate on the membrane surface, causing larger reversible fouling. Therefore, the larger the pH value is, the larger the change of the membrane flux is.

The size of the oil droplets of emulsified oil affects the degree of fouling of the membrane [32]. The effect of emulsified oil droplet size (45.27 µm, 25.53 µm, and 15.99 µm for emulsified oil) on membrane flux and membrane fouling was investigated by filtration experiments, and the results are shown in Fig. 8. As the oil droplet size decreases, the membrane flux decreases more, indicating that different oil droplet sizes cause different membrane fouling, and smaller oil droplet sizes cause more membrane fouling (Fig. 8a). In addition, as the filtration time increases, the membrane pores become clogged, and an oil layer forms on the membrane surface, which causes the membrane flux to decrease further. As shown in Fig. 8b, the graph of the membrane fouling changes shows that as the oil droplet size increases, the reversible fouling gradually increases, the irreversible fouling gradually decreases, and the total membrane fouling also shows a decreasing tendency. The strong adsorption between oil droplets and membrane pores and surfaces makes it easier for smaller oil droplets to enter the membrane pores, thus forming more serious irreversible fouling. It is consistent with the findings of Lu et al., who concluded that small organic molecules contribute more to the flux decrease during filtration.

3.2.3. Fouling of the membrane by coexistence of inorganic particles and emulsified oil

Continuous casting turbid ring water is a mixture of a large amount of iron oxide, metal dust, oil, and other impurities. In the filtration process, it greatly impacts the fouling of polyethylene porous tube membrane [4]. Therefore, when inorganic particles and emulsified oil coexist, it is necessary to conduct a detailed analysis of the microscopic morphology, membrane flux changes, and membrane fouling changes after membrane fouling.

3.2.3.1. Microstructure of polyethylene pipe membrane after fouling

After filtration of turbid ring water in which inorganic particles and emulsified oil coexist, the microscopic morphology of the outer and inner walls of the polyethylene porous filter tube membrane was characterized by scanning electron microscopy. The result shows that the polyethylene particles are not destroyed during the filtration process (as shown in Fig. 9), but only impurity particles appear in different degrees on the inner and outer walls. It can be seen from Fig. 9a that after the use of polyethylene porous filter tube membrane, the polyethylene particles on the outer wall are wrapped by a large number of impurity particles and adsorbed and deposited inside the membrane pores, which made the membrane pores reduced or even disappeared. From Fig. 9b it can be seen that a large amount of particulate matter is mainly adsorbed in the pores of polyethylene particles, indicating that impurity particles will be absorbed and retained by the membrane pore size to achieve filtration during the filtration process, which is the main reason for the decrease of the membrane flux and membrane fouling. From Fig. 9c and d it can be seen that only a small number of impurity particles are adsorbed in the pores of polyethylene particles and membrane at the inner wall of the polyethylene porous filter tube membrane after use, indicating that most of the particle pollutants are removed before reaching the inner wall of the membrane in the filtration process, indicating that the polyethylene porous filter tube has good filtration performance.

3.2.3.2. Effect of coexistence of inorganic particles and emulsified oil on the membrane fouling situation

Fig. 5 shows the membrane flux change curve and membrane fouling change during filtration of turbid ring water with the coexistence of emulsified oil and inorganic particles. From Fig. 10a it can be seen that when emulsified oil is present alone, the flux decrease is more obvious in the same filtration time compared with the filtration process

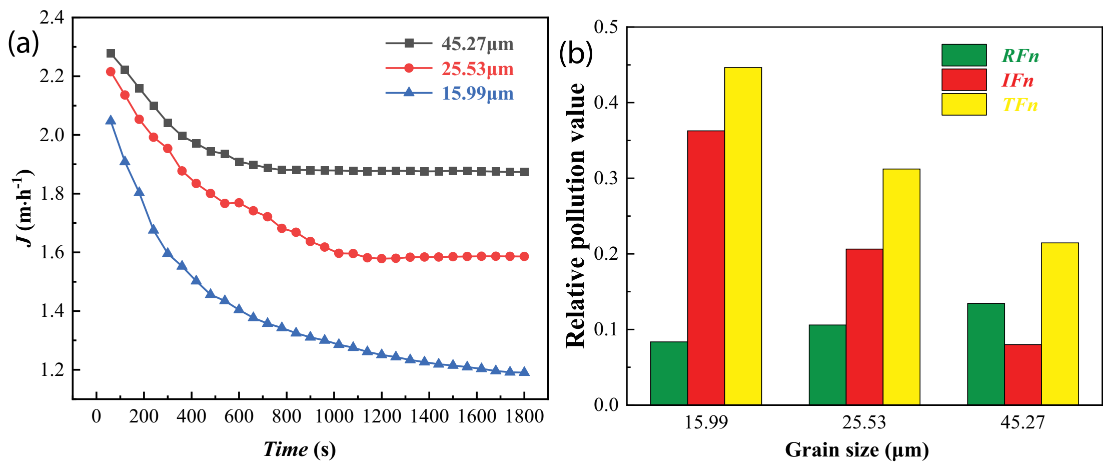

Fig. 8. Effect of different emulsified oil sizes on the fouling behavior of the membrane: (a) curve of the membrane flux and (b) fouling of the membrane.

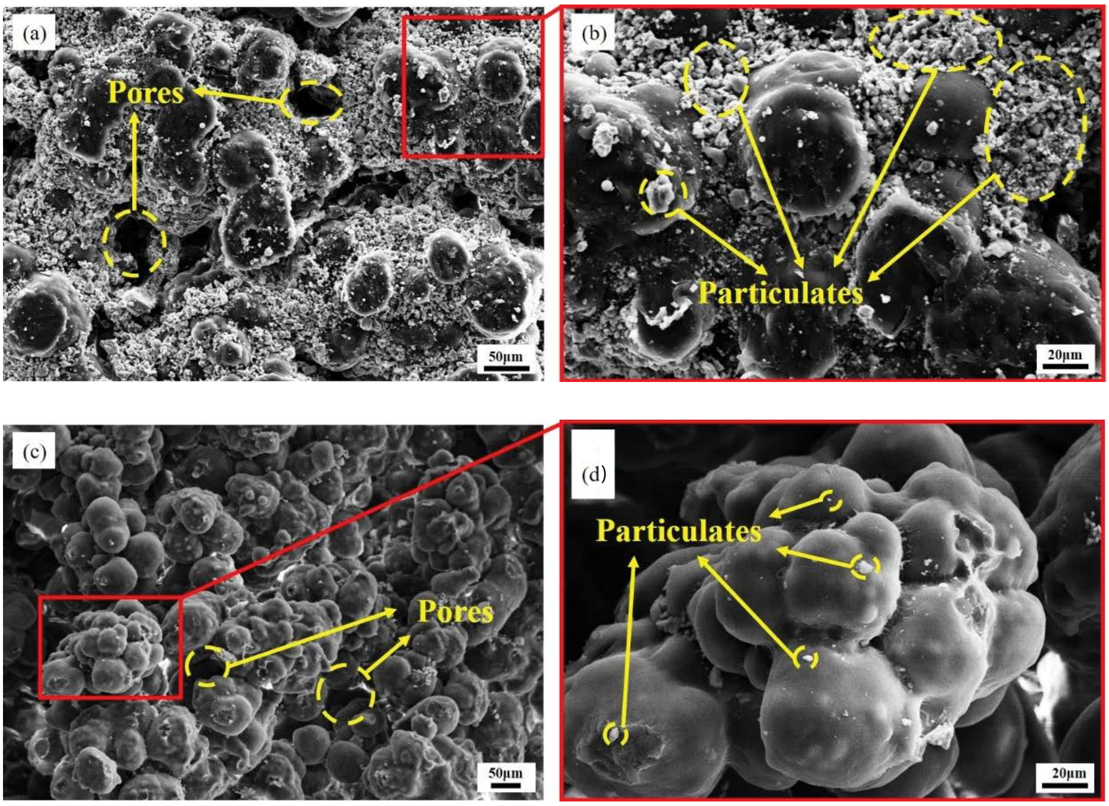

Fig. 9. Micro-SEM images of the contaminated polyethylene tube membrane (a) outer wall, (b) enlargement of figure (a), (c) inner wall, and (d) enlargement of figure (c).

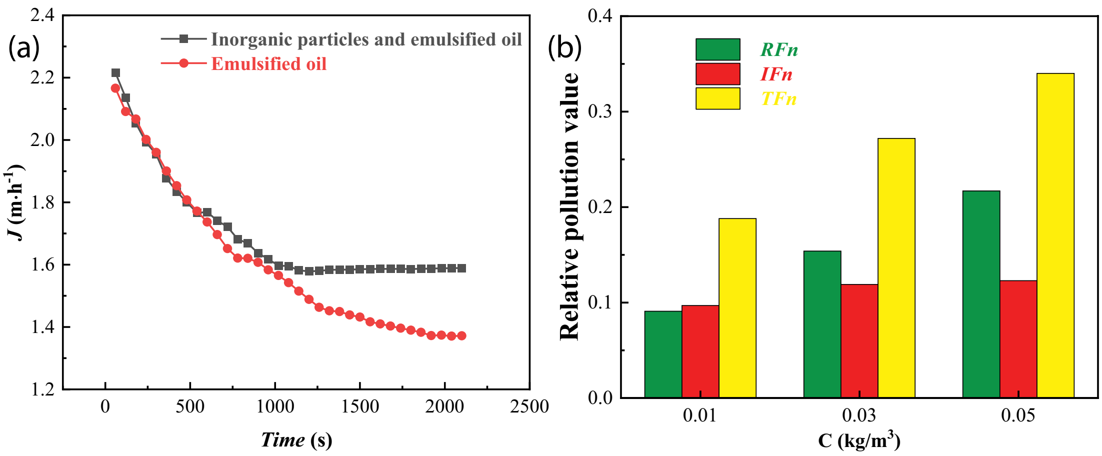

Fig. 10. Fouling of the membrane when emulsified oil and inorganic particles coexist: (a) membrane flux variation curve and (b) fouling of the membrane.

after adding inorganic particles. It is because when inorganic particles are present, the emulsified oil droplets will be adsorbed on the surface of inorganic particles to occur synergistically during the filtration process [33,34], and the emulsified oil will not form an oil membrane on the membrane surface alone. When only emulsified oil exists, the surfactant in the emulsified oil has an enveloping effect during the filtration process. It is difficult for the oil droplets to agglomerate. The intercepted oil droplets form an obvious continuous oil film on the membrane surface, which gradually reduces the effective filtration area of the membrane and leads to a more serious decrease in its membrane flux [35]. The addition of inorganic particles alleviates the membrane fouling caused by organic molecules [36]. Fig. 10b shows the membrane fouling in the filtration process of turbid ring water with different concentrations of emulsified oil (0.01, 0.03, and 0.05 kg/m^3, respectively) and inorganic particles coexisting. In the filtration process of turbid ring water, with the increase of emulsified oil concentration, the reversible fouling, irreversible fouling, and total fouling of the membrane all show an increasing trend. It is mainly because with the increase of emulsified oil concentration, the thicker the cake layer formed by the accumulation of emulsified oil and inorganic particles is on the membrane surface, and the greater the membrane resistance is. It can be seen from Fig. 10b that the vast majority of fouling is reversible fouling, accounting for between 48% and 79% of the total fouling, which can be removed by backwashing the tubular membrane with water to remove the filter cake.

4. Conclusion

In this research, porous polyethylene tube membranes for continuous casting turbid ring water purification were prepared by using ultra-high molecular weight polyethylene as raw material, their microscopic morphology was characterized, the effects of different component properties on the fouling characteristics of porous polyethylene filter tubes were investigated, and the membrane fouling process was fitted and analyzed using four classical models. The main conclusions are as follows.

- The prepared polyethylene porous filter tube was made of the accumulation of particles with different sizes of about 10–60 μm, and there were a large number of irregularly shaped three-dimensional through pores in its cross-section with the pore size of about 20–70 μm. The outer wall of the tube membrane showed a flat shape, and the inner part was more regular spherical particles.
- In the filtration process of the dispersion system containing only inorganic particles, the membrane flux decreased more as the concentration of inorganic particles increased, and at the later stage of filtration, they all showed a stable trend; the dispersion system with larger concentration was more likely to form a cake layer on the membrane surface than the dispersion system with smaller concentration, which caused larger reversible fouling, but the irreversible fouling decreased. The smaller the size of inorganic particles was, the easier it was to cross the membrane surface and enter the pore space for deposition and adsorption. Therefore, 70 μm inorganic particles caused the largest reversible fouling, and 5 μm caused the largest irreversible fouling. The flow rate had an important effect on membrane fouling; the higher the filtration flow rate was, the more the membrane flux decreased, and the reversible, irreversible, and irreversible fouling increased. The analysis shows that at the beginning of filtration, the membrane first undergoes complete pore blocking, and as the filtration process proceeds, the contamination pattern of the membrane changes and cake filtration occurs.
- In the filtration process of emulsified oil, as the size of emulsified oil decreased, the membrane flux decreased faster, and the irreversible fouling became more serious. pH value in the range of 6~10, with the increase of pH value, the zeta potential of emulsified oil decreased significantly, and the size of emulsified oil also decreased; as the pH of emulsified oil increased, the flux decreased more, and the resulting membrane fouling was greater. The model-fitting results showed that there are two main fouling models for the filtration process of emulsified oil: cake blockage and standard blockage.
- During the filtration of turbid ring water coexisting with inorganic particulate matter and emulsified oil, the membrane flux of the system with inorganic particulate matter and emulsified oil coexisting decreased less compared with the emulsified oil filtration system alone, and the addition of inorganic particulate matter was beneficial to hinder the decrease of the membrane flux. The total membrane fouling was 28.12%, and irreversible fouling was 8.95% when emulsified oil was present alone; the total membrane fouling was 30.41%, and irreversible fouling was 11.9% when inorganic particles and emulsified oil coexisted in turbid ring water filtration, indicating that the coexistence of inorganic particles and emulsified oil increased the irreversible membrane fouling and the membrane fouling was more serious because of the synergistic effect of inorganic particles and emulsified oil.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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