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The Usage of Non-woven Fabric Material as Separation Media in Submerged Membrane Photocatalytic Reactor for Degradation of Organic Pollutants in Water

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The Usage of Non-woven Fabric Material as Separation Media in Submerged Membrane Photocatalytic Reactor for Degradation of Organic Pollutants in Water

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Abstract: Photodegradation tests with continuous mode were carried out in a photocatalytic reactor containing a submerged flat sheet non-woven fabric membrane, which was able to retain the photocatalyst. The non-woven fabric membrane was made of polypropylene with a pore size of 2 μm. In this study, three operational parameters were investigated: light intensity, feed concentration, and feed flow rate. Experimental results showed no significant variation of permeate flux and TMP (transmembrane pressure) during the experimental period. The SS (suspended solid) of the permeate was non-detectable. These behaviors were due to the formation of a dynamic porous cake layer on the non-woven fabric membrane surface in our experimental condition. We also found that it was important to control the operation parameter

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mentioned above as to exactly when we wanted to maintain the steady pollutant concentration in the permeate at very low value.

Keywords: Non-woven fabric membrane, photocatalysis, photocatalytic membrane reactor

INTRODUCTION

The TiO_2 mediated photocatalytic oxidation process is currently accepted as one of the most promising technologies for water purification. In considering this photocatalytic oxidation process, a photocatalyst slurry (suspension) reactor system is developing now in our laboratory. In this process, the TiO_2 slurry (aqueous suspension) is illuminated with UV and TiO_2 catalyzes the oxidation of dissolved organic pollutants, mineralizes them to carbon dioxide, water, and mineral acids through the photogeneration of a strong oxidant such as hydroxyl radical. In the development of the slurry type photocatalytic reactor technology, the separation and reuse of the TiO_2 particle is an important issue. Many classical solid–liquid separation methods, such as the sedimentation of TiO_2 particles after suitable pH and ionic strength adjustment or coagulation with coagulant had been studied over many years (1). Besides these chemical methods, another separation method without a chemical additive is the photocatalyst slurry reactor system coupling with the membrane processes, such as microfiltration (MF) or ultrafiltration (UF), to provide a hybrid treatment for low polluted water. In this case, the membrane is used as the filtration media for the separation of the photocatalyst particle from the reaction system (2). However, this technique is an impractical application due to high membrane cost and also its operation cost. If this technique is to have a commercial future as a method for water purification, the membrane cost must be cut down or one has to find other low cost filtration material.

It is well known that non-woven fabric material is a cheaper filter material for water treatment (3). In our laboratory, as the successful experiences in developing a submerged non-woven fabric membrane bioreactor technology for wastewater treatment (4), we also tried to use the submerged non-woven fabric membrane as a filtration media for photocatalyst slurry in a photocatalytic membrane reactor (PMR) system.

Hence the purpose of this study was to investigate the filtration ability of the non-woven membrane in a submerged non-woven fabric membrane photocatalytic reactor (SNMPR) system. It was designed for the degradation of methylene blue in water with a photocatalyst slurry in this system.

METHODS

The schematic diagram of the submerged non-woven fabric membrane photocatalytic reactor system used in this study was shown in Fig. 1. The reactor was made of pyrex glass with a working volume of 22.4 L. The reactor

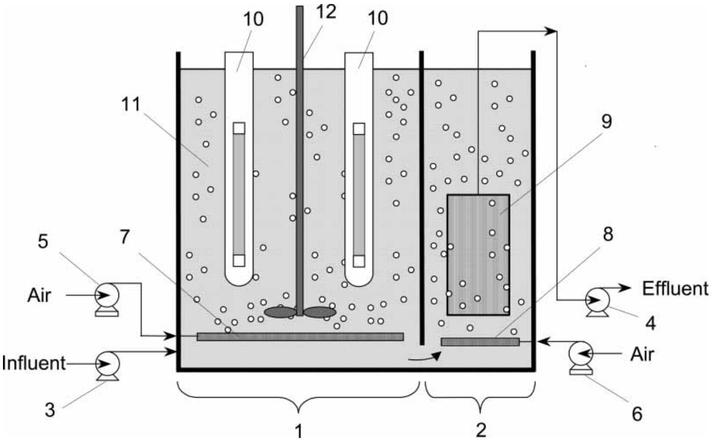

Figure 1. Scheme of submerged non-woven fabric membrane photocatalytic reactor system in this study. 1. Photocatalytic reaction region, 2. membrane separation zone, 3. influent pump, 4. effluent pump, 5&6. blower, 7&8. air distributor, 9. membrane module, 10. UVA lamp, 11. photocatalyst suspension, 12. stirrer.

was divided into two regions by a baffle: the photocatalytic reaction region with volume of 16 L and membrane separation region with volume of 6.4 L. The non-woven fabric membrane module was submerged into the membrane separation region. In each experiment, aqueous suspensions of TiO_2 containing a different amount of methylene blue was stirred at constant speed with 120 rpm and irradiated by UV light. The cover of the glass reactor was designed with inlets for air, taking samples and a glass tube holder for UV lamps. The UV radiation was performed from four black lamps of 4 W or 6 W power and a wavelength of 365 nm with light intensity of 6 mW/cm^2. Methylene blue was chemical reagent grade. The titanium dioxide photocatalyst was Degussa P-25 (70% anatase and 30% rutile, mean primary particle size 30 nm). When the Degussa P-25 photocatalyst powder was dispersed in water, because the interfacial attractive force between the titanium dioxide nanoparticle is very large, secondary aggregates were formed due to the cluster effect of the nanoparticle. The diameter of these secondary aggregates was in the range of μm (5). The concentration of the TiO_2 aqueous suspension in the reactor system was 350 mg/L. A methylene blue solution was fed by a suction pump. All of experiments were carried out at room temperature (25° C) and in the presence of air. The reaction was monitored by the absorbance of the methylene blue at 655 nm. It was measured by using a Hitachi u-2000 UV-vis spectrometer. A suspended solid (SS) analysis was carried out in accordance with Standard Method 2540.

The membrane material was a non-woven fabric polypropylene material having an average pore size of 2 μm. It was supplied by KNH Enterprise Co., Ltd, Taiwan. The membrane module included a square case and double layers of sheet-like structures fastened within the square case, wherein a space of 2 mm was formed between the double layers of the sheet-like structures as a channel for the water flow and a support structure was installed in the channel. The periphery of the square case was sealed, and only an effluent tube was connected to the channel. The surface area (filtration area) of the sheet-like structures on the two sides was about 855 cm^2 in total. The aeration units were installed at the bottom part of the reactor to supply oxygen and induce a cross flow along the membrane surface. A suction pump allows the permeate to be collected from outside to inside through the membranes.

RESULTS AND DISCUSSION

Non-woven Fabric Material as Filtration Media in PMR System

As described in the methods section, when the Degussa P-25 photocatalyst powder was dispersed in water, secondary aggregates were formed and the diameter of these secondary aggregates was in the range of μm. Hence, the main thing we are concerned with in studying the separation of the photocatalyst in our Degussa P-25 photocatalyst slurry system was the filtration phenomena of secondary aggregates of the photocatalyst particle.

For a long time, in selecting filtration media for air or liquid filtration, non-woven fabric material has been extensively used for the removal of particles with size above μm in the decontamination process (3). Gimbel el al. also found that filtration with a permeable synthetic collector (PSC), such as porous polyurethane foam or non-woven fabric material, could get much better performance in a particle collection. The enhancement of filtration efficiency with PSC is owing to the small particle deposition inside the collectors (6). Hence it is reasonable to use non-woven fabric material as filtration media for the photocatalyst slurry in photocatalyic membrane reactor system. The configurations of the photocatalytic membrane reactor system could be a batch-recirculated photocatalyst slurry system or submerged membrane photocatalyst slurry system (7). In our previous study with the batch-recirculated photocatalyst slurry system, it was found that a non-woven fabric membrane could be used as the filtration material of TiO_2 in this system (8). The experimental results indicated that we could obtain a stabilized permeate flux, around 0.1 $\mathrm{m}^3/\mathrm{m}^2$. day, and TMP was at the value of 1 to 2 kPa. The particle collection efficiency of the non-woven fabric membrane module was more than 98% during the experimental period. Based upon this experience, we will investigate the filtration characteristics of the non-woven membrane in submerged non-woven fabric membrane photocatalytic reactor system in the following section.

Filtration Characteristics of SNMPR System

Permeate Flux and TMP

No flux decline was observed in our experimental condition and the permeate flux was almost the same as the applied initial flux. Figure 2 shows TMP as a function of time in the presence of methylene blue at a different permeate flux. It was found that we can get the same steady state TMP value irrespective of the different permeate flux although a lower TMP value was observed initially and then got to higher and steady state TMP with the value of 3.3 kPa at the lowest permeate flux case.

The filtration performance of our submerged non-woven fabric membrane photocatalytic reactor system was not the same as that of conventional photocatalytic microporous membrane reactor systems found in literature. For example, Sopajaree et al. illustrated that the permeate flux showed an abrupt decrease with time and then attained a plateau in studying the integrated photoreactor-UF process assembly with methylene blue and Degussa P-25 used as the test pollutant and photocatalyst, respectively (9). Lee et al. also investigated using of a UF membrane for the separation of TiO_2 photocatalysts in drinking water treatment. They found that a noticeable permeate flux decline was observed when humic acid (4 mg/L) was added to the system (2).

Formation of Dynamic Membrane Layer on Non-woven Fabric Membrane Surface

Non-woven fabric material is composed of a random network of overlap fiber creating a multiple connected pore through which the fluid can flow.

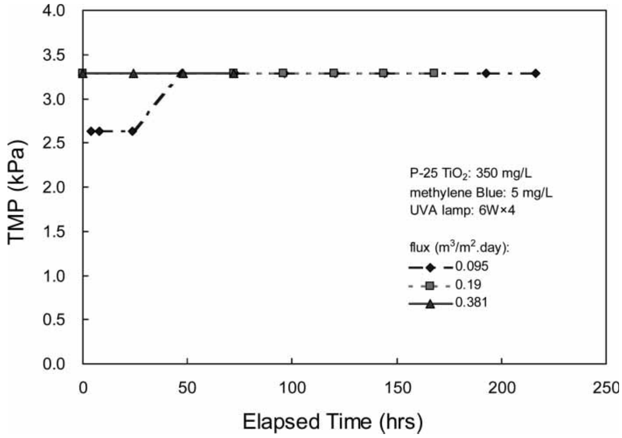

Figure 2. TMP as a function of time at different permeate flux.

The fouling tendency of this fibrous filtration material is related to the ability of the particle capture by fiber and the degree of particle re-entrainment from the filtration material (10). The major difference between microporous membranes and our non-woven fabric membrane was the pore size. The pore size of a non-woven fabric membrane used in this study was much larger than that of the microporous membrane. Hence the reason for getting a stabilized permeate flux and TMP in a large pore size non-woven fabric membrane photocatalytic reactor system could be owing to the formation of porous dynamic cake (membrane-like) layer on the non-woven fabric membrane surface. The filtration performance can be improved by a membrane self-formed dynamically through exposing on a porous body to a solution containing colloidal particles of certain hydrous oxides was first proposed by the researcher at the Oak Ridge Laboratories (11). The formation of a dynamic membrane during filtration could enable the separation of smaller particles even if the used membrane has a larger pore size. Altman et al. studied the removal of organic foulants from feed waters by a dynamic membrane formed from hydrous zirconium oxide colloidal suspension on an inexpensive non-woven flat sheet supports (12). They found that the pore size of non-woven sheet supports had little effect on the organic carbon reduction and the effluent water quality could be kept at a stable level. They suggested that it is probably due to the filtration and separation ability of a dynamic membrane layer on the non-woven membrane surface. Cai et al. also studied the separation performance of a dynamic membrane formed with MnO_2 on a porous polyethylene tube support (13). They found that it could get UF or MF performance using polymer support with the pore size in the range of 5–20 μm.

The proposition that a dynamic membrane layer formed with TiO_2 particles on a non-woven fabric membrane surface in our non-woven fabric membrane photocatalytic reactor system could be confirmed by the SS data of the photocatalytic reactor system and permeate. Figure 3 illustrated a variation of SS at a different feed flow rate. Initially the SS of the photocatalytic reactor system was 350 mg/L; however, it was found that the steady state SS of the photocatalytic reactor was less than 350 mg/L especially at the high flow rate during the experimental period. These results indicated that the photocatalyst deposited on a non-woven fabric membrane surface and formed a dynamic membrane layer. The good filtration performance of this dynamic membrane layer could be further verified by the SS of the permeate. It was found that the SS of the permeate was generally not detectable during the experimental period. The initial TMP increased with time in the lowest permeate flux case shown in Fig. 2 indicated that some TiO_2 particles penetrated and deposited inside the pore of the non-woven fabric membrane prior to the formation of the dynamic membrane layer on the non-woven fabric membrane surface. However, the permeate flux was not affected by this little pore blockage due to the larger pore size of our used non-woven fabric membrane.

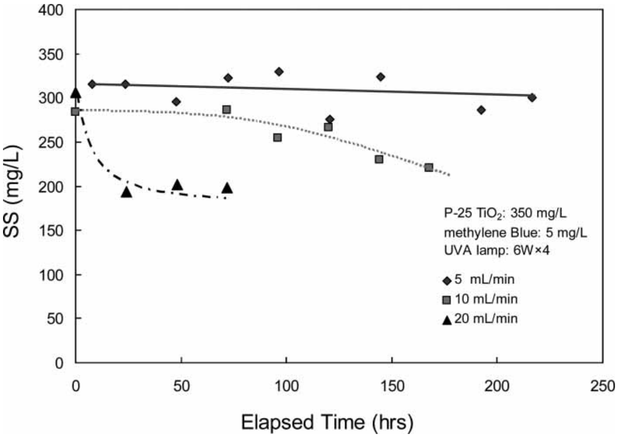

Figure 3. Variation of SS at different feed flow rate.

Performance in Continuous SNMPR System

Effect of Light Intensity

The light intensity effect on the photocatalytic degradation at constant methylene blue concentration (5 mg/L) and fixed feed flow rate (5 mL/min) in continuous SNMPR system was shown in Fig. 4. It can be seen that the lower UV absorbance value was observed at a higher light intensity. This means that a great percentage of methylene blue was photocatalytically degraded at a higher light intensity in our experimental condition. It also

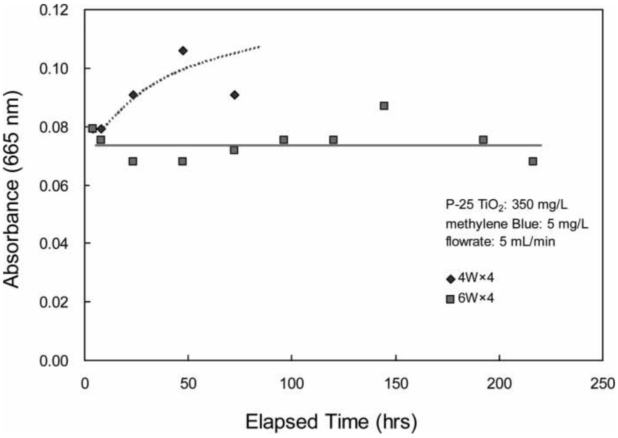

Figure 4. The UV absorbance with different light intensity at constant methylene blue concentration (5 mg/L) and fixed feed flow rate (5 mL/min).

appeared that the UV absorbance value increased with increasing time in lower light intensity. This result indicated that the light intensity was too weak and a minor amount of methylene blue was photodegraded in such case; therefore, the UV absorbance value increases with increasing time.

Effect of Feed Flow Rate

Figure 5 shows the UV absorbance value of the permeate versus the elapsed time at two feed flow rate. It can be seen that the absorbance value of the permeate was low and almost constant at 5 mL/min and UV absorbance value of permeate increase with increasing time at 50 mL/min. The photodegradation efficiency of the photocatalyst system was constant at a fixed radiation intensity and photocatalyst concentration. Decreasing the feed flow rate means that increasing the residence time of methylene blue and thus have enough time for photodegradation. The result in Fig. 5 demonstrated that the degree of methylene blue photodegraded was almost complete at 5 mL/min in the entire elapsed time. However, the methylene blue concentration was too high to degrade completely at 50 mL/min case.

Effect of Feed Concentration

Figure 6 shows the time profiles of photocatalytic degradation under different methylene blue concentrations in a reaction system at fixed light intensity and feed flow rate. For comparison explicitly, we investigated a variation of methylene blue concentration instead of using the absorbance value. It is clear from Fig. 6 that the methylene blue concentration in the reaction system is very low and almost no variation at feed

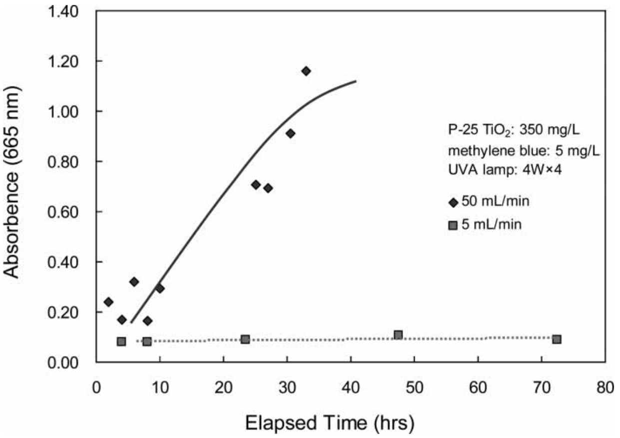

Figure 5. UV absorbance of permeate versus elapsed time at two feed flow rate.

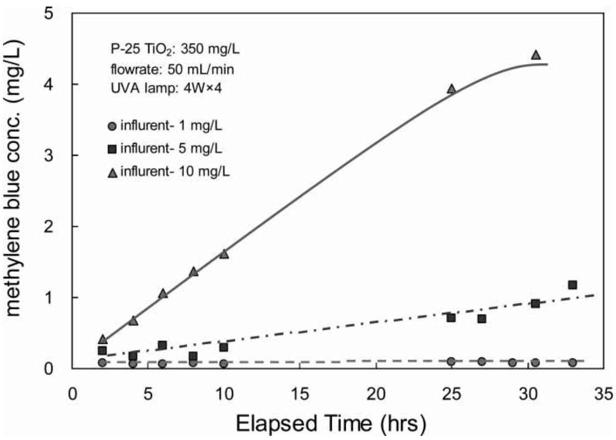

Figure 6. The time profiles of photocatalytic degradation under different methylene blue concentration at fixed light intensity and feed flow rate.

concentration of 1 mg/L. However, methylene blue concentration in the reaction system increase with increasing feed concentration at two other feed concentrations and increasing more with a higher feed concentration. This increasing of methylene blue concentration in the reaction system is due to the limiting effect of applied light intensity of our experimental system.

In a continuous photocatalytic water treatment, it is important that the pollutant must be mineralized completely or a steady state pollutant concentration in the permeate kept at minimum value. From the above data, we could see that the flow rate and feed concentration set at a suitable lower value as possible was necessary.

CONCLUSION

1. Non-woven fabric membrane could be used as a filtration material for a photocatalyst slurry on a submerged membrane photocatalytic reactor system.
2. Experimental results showed no significant variation of permeate flux and trans-membrane pressure during the experimental period. The SS of permeate was non-detectable. These behaviors were due to the formation of a dynamic porous cake layer on the non-woven membrane surface.
3. In continuous photocatalytic water treatment, the flow rate and feed concentration must set at a suitable lower value as possible, then the steady state pollutant concentration in the permeate could be kept at a minimum value.

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