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Stability study of PVDF/TiO_2 dual layer hollow fibre membranes under long-term UV irradiation exposure

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ABSTRACT

Photocatalytic membrane has received overwhelming attention in recent years and has been considered as one of the most promising advanced oxidation processes. In such system, the photocatalytic membranes are constantly exposed to UV irradiation and the effects of UV irradiation on the structure and long-term stability of polymer membranes are still questionable, and therefore, it deserves to be investigated. This study focuses on the stability of the TiO_2/PVDF dual layer hollow fiber (DLHF) membranes against photocatalytic reactions for photocatalytic-membrane process. 3 wt% of TiO_2 nanoparticles loading in DLHF membranes was evaluated under 8 W UVA in 30 days. The membrane stability was characterized by observing the changes in Fourier transform infrared spectroscopy (FTIR), atomic force microscopy (AFM), scanning electron microscope (SEM) and tensile strength analysis. The results revealed that some differences in FTIR peaks between new membranes and used membranes were found, suggesting that UV irradiation has some effect on the stability of the TiO_2/PVDF membrane for 30 days exposure. The tensile strength of used membrane showed a moderate decrease with increasing UV exposure time, resulting in negative impacts on the membrane overall stability. This work is particularly importance to evaluate the sustainability of polymeric membrane, which has considered as heart of photocatalytic membrane reactor for wastewater treatment process.

1. Introduction

Recently, membranes immobilized with TiO_2 nanoparticles have gained interest in hybrid photocatalytic membrane, as one of advanced waste water treatment process. These hybrids photocatalytic membrane will have a synergy of photocatalytic and filtration technologies resulting in a very powerful system, with the membrane having the simultaneous task of supporting the photocatalyst as well as acting as a selective barrier for the species to be degraded.

In photocatalytic system, the main factor that influenced the photocatalytic performance is catalyst and UV light. During the operation, the photocatalytic membranes are constantly exposed to UV irradiation. The method for immobilized TiO_2 nanoparticles also influences the stability of the membrane itself. As reported by previous researchers, the deposited TiO_2 nanoparticle [1] at the outer surface of membrane are more stable under long time UV irradiation compared to immobilized within membranes [2] because of deposited TiO_2 would covered the membrane which hindered it directly exposure to UV light. Meanwhile, potential damage to the TiO_2 within membranes is highly expected due to the direct exposure of membranes to UV light. The depth of UV penetration in photocatalytic process has been explored by many researchers as listed in Table 1. The penetration of UV light to membranes would depend on porosity and pore size. It is due to the reaction kinetics were controlled by the kinetics of product transport through the porous structure that entraps the photocatalyst.

To date, most of researchers are more focused on the photocatalytic performance by using different type of photocatalyst compared to stability of membrane itself [6,7]. Mostly, in their experiments, the irradiation period was limited within 12 h [8–10]. This limitation time is not representing the long term stability of the membrane itself. Only one publication has been found to be directly related to the evaluation of stability of flat sheet membranes under photocatalytic process [11]. They reported that PVDF, PTFE and PAN shows the 30 days of UV irradiation shows the greatest stability.

On the other hand, Ong et al. reported that the outer surface layer of membrane in hollow fiber configuration was cracks and fractures after a direct exposure to 120 h UV light. The membrane structure

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Table 1
Depth of UV penetration in photocatalytic process.

Sample	Depth of UV penetration (μm)	References
TiO_2 film	0.6	Danion et al. [3]
TiO_2 film	1–2	Choi et al. [4]
Immobilized TiO_2	Depend on porosity & pore size	Tahiri et al. [5]

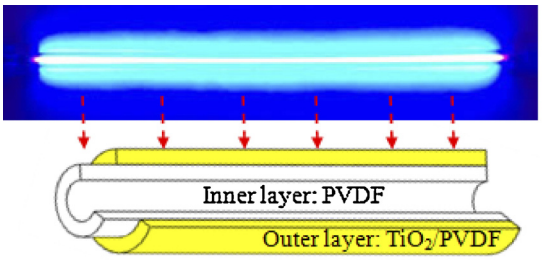

Fig. 1. UV irradiation on dual layer hollow fiber membranes.

was collapse when the exposure has been extended to 250 h and after that, turning it into powder form. They claims that membrane was degraded at high UV exposure time based on the reduction of membrane wall thickness and lower carbon and flouride contents [12]. However, the finding can be argued as the membrane in the work was irradiated very close to the membrane in air environment, which is contrary to real condition of photoreactor applied for water treatment. It is expected that the results would be different if the test was conducted in water.

To our best knowledge, there have been no systematic studies that have reported on the long-term effect of UV irradiation on dual layer hollow fiber membranes in photocatalytic membrane reactor as shown in Fig. 1. In this article, we will first report a systematic investigation on the limits and stability of the polymeric membranes and/or alternative membrane materials against photocatalytic reactions for water treatment applications. Based on our previous findings, dual layer hollow fiber membranes are more effective in nonylphenol degradation compared to single layer hollow fiber membranes due to the better dispersion of TiO_2 on the outer membrane surface [13]. However, investigation on the stability of dual layer hollow fiber membranes exposed to UV irradiation has not been done. Therefore, this study is important to be carried out in order to evaluate the effect of UV irradiation during photocatalysis on the polymeric membranes.

2. Experimental

2.1. Membrane preparation

PVDF and TiO_2 were dried in a 50 °C vacuum oven for 24 h to remove moisture prior to dope preparation. To prepare the outer dope solution, 3 wt% of TiO_2 and 82 wt% of dimethylacetamide (DMAc) were mixed and stirred inside a Scott bottle with an overhead stirrer at 400 rpm for 24 h. After the TiO_2 mixture became a well-dispersed solution, 15 wt% of PVDF was gradually added. The similar technique was applied for preparation of inner dope solution. The inner dope composition of PVDF and DMAc are 18 wt% and 82 wt%, respectively. Then, each solution was degassed by using an ultrasonic bath system at ambient temperature over night prior to spinning. The spinning dope mixture was extruded using a triple orifice spinneret to form dual layer hollow fiber membranes as stated detail elsewhere [14].

2.2. Stability test

Experimental works using a batch submerged membrane photoreactor were conducted to evaluate the stability of dual layer hollow fiber membranes in photocatalytic process. 6 bundles of 15 hollow fibres with approximate length of 23.5 cm (total effective membrane area: 248 cm^2) was potted into PVC tube using epoxy resin (E-30CL Loctite® Corporation, USA). The module was then left at room temperature for hardening before its protruding parts were cut and placed into a rounded polystyrene container to complete the module preparation. The rounded polystyrene was covered with aluminium foil in order to control the UV light.

The prepared module was then fitted in the ultraviolet A (UVA) photoreactor system as shown in Fig. 2. A 100 ppm of feed solution was prepared by dissolving 1 g nonylphenol (NP) in H_2O:acetonitrile (9:1) [15] with 10 L of volume. The solution was stirred at 298 K in the dark for 1 h (adsorption equilibrium) prior to UVA photocatalytic degradation. The reaction solution was moderately stirred using magnetic stirrer in order to prevent pollutant deposition at the bottom of the reactor.

The external beaker surface was covered with aluminium foil whose reflecting surface was directed in the inner side. A Philips lamps (UVA radiation, 8 W) emitting radiation in the 365 nm of wavelength was used to irradiate the immobilized TiO_2 in membrane sample. The lamp was placed in the middle of the beaker. The UV light intensity exposed to each bundles of hollow fiber membrane would be assumed similar due to the position of each membrane to UV light is 4.5 cm of distance. A bundle of membranes was taken out for further analysis for 5 day within 30 days of experiment.

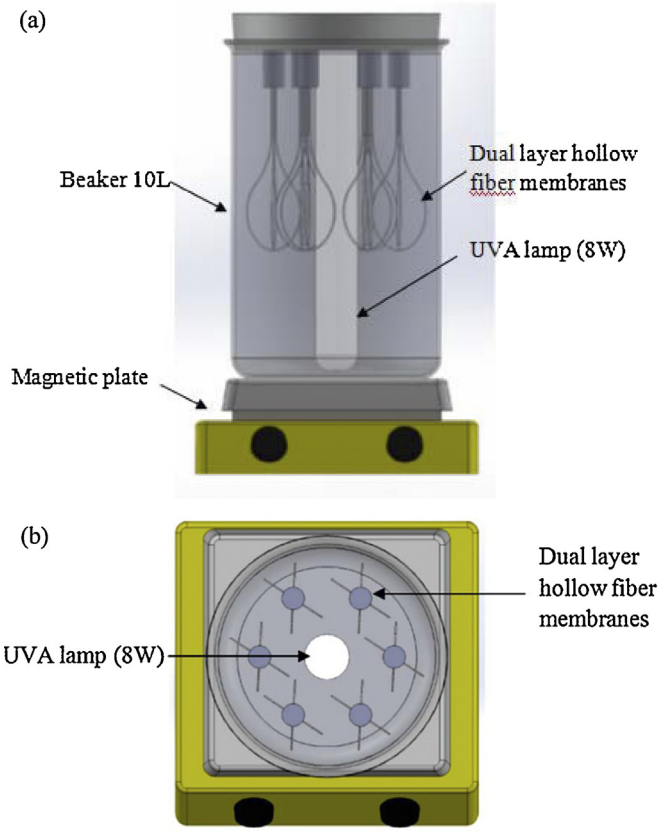

Fig. 2. Schematic diagram of submerged photocatalytic membrane apparatus (a) front and (b) top view.