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Effects of hydrophilic surface macromolecule modifier loading on PES/O-g-C₃N₄ hybrid photocatalytic membrane for phenol removal

Noor Elyzawerni Salim^{a,b}, N.A.M. Nor^{a,b}, Juhana Jaafar^{a,b,*}, A.F. Ismail^{a,b}, M.R. Qtaishat^c, T. Matsuura^d, M.H.D. Othman^{a,b}, Mukhlis A. Rahman^{a,b}, F. Aziz^{a,b}, N. Yusof^{a,b}

^a Advanced Membrane Technology Research Centre (AMTEC), Universiti Teknologi Malaysia, 81310 UTM Skudai, Johor, Malaysia

^b Faculty of Chemical and Energy Engineering (FCEE), Universiti Teknologi Malaysia, 81310 UTM Skudai, Johor, Malaysia

^c Chemical Engineering Department, The University of Jordan, Amman 11942, Jordan

^d Industrial Membrane Research Laboratory, Department of Chemical Engineering, University of Ottawa, Canada

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ABSTRACT

Hydrophilic surface modifying macromolecules (LSMM) modified polyethersulfone (PES) based photocatalytic membranes have been successfully prepared. This hybrid photocatalytic membrane applying oxygen-doped graphitic carbon nitride (g-C₃N₄) as a photocatalyst was successfully fabricated via phase inversion technique in flat sheet form at ambient temperature. The potential of LSMM as membrane modifier was explored in detail under various loadings (1–5 wt%). The results show that the LSMM addition successfully increased the membrane hydrophilicity which may consequently prevent the membrane from comprehensive fouling. More appearance of g-C₃N₄ on the membrane surface was observed by Scanning Electron Microscopy (SEM) and Atomic Force Microscope (AFM) analyses upon the LSMM addition. However, the trend tended to decline at the loading beyond 4 wt% of LSMM. At 4 wt% of LSMM, the PES/g-C₃N₄ membrane successfully decreased phenol concentration up to 35.78% and rejected 14.73% of phenol. From the water flux result, the application of LSMM in water treatment application was hindered by the flux deterioration but a considerably high flux for an ultra-filtration membrane. The results indicate that the introduction of LSMM in the PES/g-C₃N₄ hybrid photocatalytic membrane showed the great potential of LSMM as a membrane surface modifier for photocatalytic activities and membrane separation performance.

1. Introduction

Photocatalysis or photocatalytic has recently become a synonym terminology in water treatment technology. The use of photocatalyst is necessary to make the water treatment by photocatalytic process realistic as when it is irradiated with light, it induces some chemical reactions and performs decomposing ability if the optimum conditions are achieved. Among all photocatalysts currently available in the market, graphitic carbon nitride (g-C₃N₄) has been intensively investigated due to its various excellent merits in response to visible light, high photochemical stability, and easily modified electronic structure [1,2]. Immobilisation technique of g-C₃N₄ has been extensively used in membrane fabrication for photocatalyst. This technique has become more selective compared to the suspended technique of g-C₃N₄ powder in solution because the technique tends to cause the catalysts to separate from the treated water and thus an additional filtration method is required to separate the catalyst from the treated feed after treatment,

which therefore incurs higher operating costs.

In the case of immobilisation, when g-C₃N₄ is embedded in the membrane, it reduces the photocatalytic active surface area as g-C₃N₄ tends to scatter in the polymer membrane. Agglomeration of g-C₃N₄ is also one main issue in which the pure g-C₃N₄ has blocky morphology due to agglomerate lamellar structure [3]. This stimulates g-C₃N₄ to agglomerate when blended in the membrane polymer. These deficit properties of g-C₃N₄ reduce the removal efficiencies of organic pollutants, which become the main obstacle to be solved in the system. In order to enhance the photoactivity of g-C₃N₄, modified g-C₃N₄ by oxygen doping was introduced to the photocatalytic membrane system. Oxygen doping increases the light radiation absorption and pollutant adsorption on g-C₃N₄ because of the oxygen-bearing groups overwhelm the recombination of electron-hole pairs [4,5]. Above all issues, this generates the idea to propose a study on how to increase the active surface area and exposure of the photocatalyst that to be embedded onto the membrane support to enhance the removal efficiencies of an

* Corresponding author.

E-mail address: 20juhana@utm.my (J. Jaafar).

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organic pollutant in wastewater.

Submerged membrane photocatalytic reactor (SMPR) is the latest technology where the composite membrane-based filtration process is coupled with the photodegradation process in one single unit. In SMPR, like the classical photoreactors, the catalyst should be immobilised in a membrane or it can be suspended in the reaction mixture. SMPR is a hybrid reactor in which photocatalysis is coupled with a membrane separator that acts as a simple barrier for the photocatalyst and a selective barrier for the molecules to be degraded. There are many engineered SMPR configurations have been designed so that it can be easily scaled up to meet the process requirement. SMPR has many advantages such as the control of residence time of molecules in the reactor, confining of the photocatalyst in the reaction environment by means of the membrane, and a continuous process with simultaneous photocatalysis and separation [6]. For immobilised photocatalysts MPR, the photocatalytic reaction takes place either on the membrane surface or within the membrane pores. The type of photocatalytic membrane used depends on the target pollutant size and the final quality of water required. The membrane is useful for separation when the size of target particle is 0.1–5 μm for microfiltration (MF) [7], while ultrafiltration (UF) and nanofiltration (NF) are for smaller size of target particles [8,9]. In order to enhance the employment of photocatalytic process, it is necessary to improve the separation and recovery of $\text{g-C}_3\text{N}_4$ catalysts from treated water. Therefore, immobilisation of catalyst into the membrane matrix can be considered as a potential technique to resolve the catalyst pollution problem.

For photocatalysis, the catalyst must be exposed to a light source and in contact with the pollutant. The support must be configured to efficiently route the pollutant to the illuminated zone and at the same time maintain a high-water flow rate to ensure good mixing without significantly increasing the system pressure, which at the end may increase the operating cost [10]. Recently, attention has been placed on improving the performance and feasibility of immobilised photocatalyst into/onto membrane. Nor et al. (2016) proposed a photocatalytic TiO_2 nanocomposite membrane prepared by hot pressing technique to enhance the contact between target pollutant and photocatalyst. However, the technique reduced the membrane flux value at high hot-pressing temperature [11].

Another approach was introduced by Dzinun et al. (2015) by fabricating a dual-layer hollow fibre membrane. Even though the technique did not achieve 100% degradation via suspended PMR as other literature, this technique successfully increased the photocatalytic degradation of nonylphenol compared to studies where TiO_2 catalyst was embedded in a single-layer hollow fibre membrane [12]. However, despite exhibiting improved photocatalytic performance, the technique reduced the water flux and required advanced facilities and equipment which might involve higher cost for membrane fabrication. Other than losing catalyst active surface area compared to that of the suspended catalyst technique, this technique also encountered fouling issues, which is a major drawback of PMR due to the separation process. Fouling may lead to flux reduction and might render the photocatalyst ineffective. Enhanced membrane hydrophilicity could overcome fouling issues. Mohamed et al. (2015) proposed an enhanced hydrophilic N-doped TiO_2 nanorods in regenerated cellulose membrane matrix which exhibited good antifouling properties and enhanced photocatalytic performance compared to those of the unmodified membranes [13].

In general, the membrane top skin layer governs the performance of a separation process. The surface deposition of contaminants from feed solutions is also affected by the surface properties of the membrane leading to reduction of the membrane performance, especially in a long-term operation [14]. In the membrane literature, to improve transport properties of polymeric membranes as well as their chemical resistance (i.e., solvent resistance, swelling, and fouling resistance), various membrane modification techniques have been developed including physical, chemical, or bulk modification (i.e., polymer blends) [15]. There are several methods to fabricate composite membranes

with a dense or porous thin film, such as solution coating or dip coating, interfacial polymerisation, graft polymerisation, lamination, plasma polymerisation or plasma deposition, and the use of surface modifying macromolecules (SMMs). Many of the surface modification methods listed above are complicated and require at least one additional step for membrane fabrication. One of the simplest methods is to introduce active additives that can migrate to the air/film interface and change its chemistry while leaving the bulk properties intact. This can be done by surface modifying macromolecules [16,17]. According to this method, membranes can be prepared by the phase inversion technique in only one casting step employing a polymer solution. It must be pointed out that various parameters affect the host polymer or polymer additive migration towards the air/membrane interface [18].

Surface modifying macromolecules (SMMs) are polymers tailor made to be compatible with a base polymer by simple blending method. The blending method of SMM with casting solution has been successfully implemented by a few researchers [19,20]. During casting, SMM with lower surface energy compared to that of the base polymer tends to migrate to membrane surface to minimise the total free energy of the system. The modified membrane surface becomes either more positively or negatively charged, resulting in more hydrophobic or hydrophilic depending on the type of SMM added. Solvent evaporation time and SMM content in the casting solution towards the SMM migration are the important parameters that need to take into account. SMM is oriented perpendicularly to the air interface, exposing these parts to air. Migration and conformational rearrangement of SMM are kinetic processes that require certain amount of times for completion. The incorporation of the SMM additive would result in the surface migration and concentration of SMM to the topmost layer of the membrane surface.

It is proven in the literature that an addition of photocatalyst into the polymeric membrane resulted in scattered photocatalytic particles all over the membrane structure. This phenomenon relatively reduces the photocatalytic surface area when the photocatalytic particles doped on the bottom of the membrane are prevented from receiving lights. As far as the subject is concerned, none of the studies investigated on how to improve photocatalytic distribution in membrane surface by blending process. Thus, the aim of this study was to improve the function of $\text{g-C}_3\text{N}_4$ surface via chemical treatment and modification by incorporating this material with a specific additive, which was LSMM that can promote $\text{g-C}_3\text{N}_4$ in absorbing more lights to improve photocatalytic activity and hence have excellent membrane separation performance.

2. Experimental procedures

2.1. Materials

The commercial polyethersulfone (PES) powders was purchased from Solvay Veradel®, US. *N*-Methyl-2-pyrrolidone (NMP) (Analytical Grade, RCI Labscan Limited, Thailand) which was employed without further purification was used as the main component for the membrane fabrication. Meanwhile, for the preparation of oxygenated graphitic carbon nitride ($\text{g-C}_3\text{N}_4$), melamine ($\text{C}_3\text{H}_6\text{N}_6$) was purchased from Sigma Aldrich and hydrogen peroxide (H_2O_2 , analytical grade, 30%) was purchased from Merck Milipore. Meanwhile, Hydrophilic surface modifying macromolecules (LSMM) was supplied by University of Jordan (molecular weight, MW, ~ 4050 g/mol) and phenol powders for the synthetic wastewater for photocatalytic testing was purchased from QRec.

2.2. Fabrication of $\text{g-C}_3\text{N}_4$ photocatalyst

$\text{g-C}_3\text{N}_4$ photocatalyst was synthesised according to our previous study [21]. The preparation of $\text{g-C}_3\text{N}_4$ involved two stages which were the preparation of graphitic carbon nitride ($\text{g-C}_3\text{N}_4$) by condensation

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