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Research Paper

Photocatalytic reusable membranes for the effective degradation of tartrazine with a solar photoreactor

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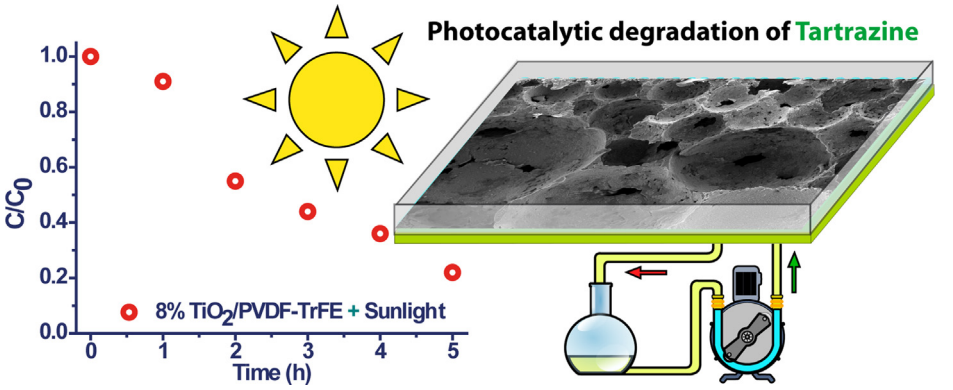
HIGHLIGHTS

- TiO_2 nanoparticles were immobilized on PVDF-TrFE for photocatalytic degradation of tartrazine.
- A solar photoreactor was used for the photocatalytic experiments.
- The dependence of tartrazine concentration on photocatalysis efficiency was addressed.
- The light intensity and the reusability of the of the TiO_2/PVDF-TrFE membranes was tested.

GRAPHICAL ABSTRACT



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ABSTRACT

Recalcitrant dyes present in effluents constitute a major environmental concern due to their hazardous properties that may cause deleterious effects on aquatic organisms. Tartrazine is a widely-used dye, and it is known to be resistant to biological and chemical degradation processes and by its carcinogenic and mutagenic nature. This study presents the use of TiO_2 (P25) nanoparticles immobilized into a poly(vinylidenefluoride–trifluoroethylene) (P(VDF–TrFE)) membrane to assess the photocatalytic degradation of this dye in a solar photoreactor. The nanocomposite morphological properties were analyzed, confirming an interconnected porous microstructure and the homogeneous distribution of the TiO_2 nanoparticles within the membrane pores. It is shown that the nanocomposite with 8 wt% TiO_2 exhibits a remarkable sunlight photocatalytic activity over five hours, with 78% of the pollutant being degraded. It was also demonstrated that the degradation follows pseudo-first-order kinetics model at low initial tartrazine concentration. Finally, the effective reusability of the produced nanocomposite was also assessed.

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1. Introduction

Water pollution caused by organic chemicals from textile, paper, plastic, leather, food, and mineral processing industries is a growing problem [1,2], mainly caused by the presence of synthetic dyes in effluents. Estimates show that approximately 10–15% of dyes used in the industry will end up in wastewaters after the manufacturing process [3,4]. Most of these dyes possess complex and stable molecular structures, making them resistant to conventional biological and chemical degradation processes [5,6]. The presence of these dyes in effluents compromises water properties, such as color, pH, sunlight penetration, and chemical oxygen demand, and important ecological impacts as most of these dyes are carcinogenic and genotoxic – associated with oxidative stress [7,8]. Among these dyes, tartrazine (C.I. Acid Yellow 23, AY23) was selected as a model pollutant for the present work. Tartrazine is an azoic dye widely used in the textile, cosmetics, pharmaceutical and food industry [9,10]. In the last decades, many works have reported about tartrazine hazards, identifying its potentially deleterious effects, such as food allergies, mutagenic, carcinogenic and phototoxicity [11–15].

In this context, the development of low-cost and efficient advanced water treatment technologies are urgently needed. There has been significant research on applying photocatalysis for water treatment due to their efficient degradation of organic compounds [16,17]. Photocatalytic reactions allow for complete degradation of organic pollutants into small and harmless species, without using chemicals, thus avoiding sludge production and disposal. Titanium dioxide (TiO_2), a well-known semiconductor, has been applied in air purification, solar energy conversion, and wastewater treatment due to its high photocatalytic activity, low cost, low photo erosion, non-toxicity, and excellent chemical and thermal stability under illumination [18]. In wastewater treatment processes, TiO_2 nanoparticles are used as a slurry system due to the large surface area of the catalysts, which allows high photocatalytic efficiency [19]. Nevertheless, the filtration to separate and recycle the TiO_2 nanoparticles suspended in the treated water significantly increases the running cost and produces collateral pollution. This problem has become a limiting factor in the practical application of photocatalytic slurry systems. To prevent secondary pollution and to allow the reusability of photocatalytic materials, numerous works have been devoted to TiO_2 nanoparticle immobilization into substrates, including glass, zeolites, ceramic particles and polymers, among others [16,20–23]. These materials can be produced by techniques such as sol-gel, electrochemical anodizing, chemical deposition, photo-assisted deposition, solvent casting, grafting, dip coating, electrospinning have been used [20,21,24–27].

Further, together with photocatalytic treatments, membrane processes have also been increasingly applied to treat waste effluents from industry. Membrane separation processes have shown to be competitive with other separation processes regarding energy costs, material recovery, environmental friendliness, and for being simple to integrate into processes for selective removal of impurities [28]. However, this technique just concentrates pollutants, not destroying them. It can be argued that the coupling of a photocatalytic reaction with a membrane separation process could take advantage of the synergy of both technologies. This combination can result in a robust system, with the membrane having the dual task of supporting the photocatalyst as well as acting as a selective barrier for the species to be degraded [2,29]. Membrane separation may be combined with a photocatalysis process in what is called a photocatalytic membrane reactor. In such reactor, the catalysts could be immobilized on the membrane surface or suspended in water [30]. However, most of the works reported concerning photocatalysis use TiO_2 suspended in water.

Many of the most used membranes are based on polymeric materials, mostly for being mechanically stable, chemically inert, inexpensive and easily accessible [31]. The first application of a polymer as a TiO_2 substrate for photocatalytic applications dates back to 1995, where TiO_2 particles were immobilized into a polythene film [32]. In the next years, many other polymers such as poly(vinylidenefluoride), polystyrene, polyethylene glycol and polyamide12, among others, were tested [21,33,34]. Poly(vinylidenefluoride–trifluoroethylene) P(VDF–TrFE) allows the production of membranes with controlled porosity and pore size [35–37]. Its favorable physicochemical properties are suited for photocatalytic application as it shows excellent UV resistance [38], which is paramount to materials continuously exposed to sunlight. Furthermore, it possesses a chemical, mechanical and thermal resistance, related to the stable C–F bonds of the polymer chain [39]. Additionally, this nanocomposite has displayed a remarkable photocatalytic activity in the degradation of methylene blue [37,40].

The principal goal of this work is to demonstrate the suitability and reusability of photocatalytic TiO_2 nanoparticles immobilized on a P(VDF–TrFE) porous membrane in the degradation of tartrazine in a solar photoreactor. Several studies have reported about tartrazine photocatalytic degradation [41–44]. However, few of them focus on the solar radiation and using immobilized catalysts.

2. Experimental

Poly(vinylidene fluoride-trifluoroethylene), (P(VDFTrFE)) 70/30 was purchased to Solvay (Belgium). Titanium dioxide (TiO_2) nanoparticles (P25-Aeroxide), with a surface area ranging from 35 to 65 m^2/g (manufacturer datasheet), were acquired to Evonik Industries AG. Tartrazine (M = 534.36 g/mol) also known as Yellow 23 (Fig. 1a), with the chemical formula $C_{16}H_9N_4Na_3O_9S_2$ and a maximum absorption at the wavelength of 427 nm (Fig. 1b) was purchased to ACROS organics (USA).

2.1. Production of TiO_2/PVDF-TrFE nanocomposite membranes

The poly(vinylidene difluoride)-co-trifluoroethylene (P(VDF-TrFE)) membranes containing titanium dioxide P25 were prepared by solvent casting according to [37]. This filler concentration was selected based on previously reported works [37] to reduce nanoparticles aggregates, detachment from the substrate and preserve the required polymer mechanical properties. Briefly, as represented in Fig. 2, 0.86 g (8 wt.%) of TiO_2 nanoparticles was added to 90 ml of N, N-dimethylformamide (DMF, Merck) and placed in an ultrasound bath for four hours to achieve a good dispersion of the nanoparticles. Then, 10 g of P(VDF-TrFE) was added to the solution, reaching a concentration of 10 wt.% polymer, and kept under magnetic stirring until complete dissolution. Finally, the solution was placed in a glass support to allow solvent evaporation, at room temperature.

After complete evaporation of the solvent, a membrane with the same dimensions of the photoreactor tank surface (38 cm length × 12 cm wide cm) was precisely cut out.

2.2. Characterization of TiO_2/PVDF-TrFE nanocomposite membranes

The microstructure of the produced nanocomposite membranes was evaluated by scanning electron microscopy (SEM). The samples were coated during 30 s with a thin gold layer and analyzed with a Quanta 650 SEM (FEI). Energy dispersive X-ray spectroscopy (EDX) was assessed with an INCA 350 spectrometer (Oxford Instruments).