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Modification of polypropylene filter with metal oxide and reduced graphene oxide for water treatment

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Abstract

A hydrothermal method for the synthesis of reduced graphene oxide/titanium dioxide filter (RGO/TiO_2) and reduced graphene oxide/zinc oxide filter (RGO/ZnO) by using polypropylene (PP) porous filter is reported. Field emission scanning electron microscopy illustrated that the nanoparticles were uniformly distributed on the reduced graphene oxide nanosheets. Flexural tests showed that the physical properties of the modified filters have greater strength than the original filter. Thermogravimetric analysis revealed that the thermal property of the modified filters is the same as that of the original filter. Under a halogen lamp, the modified filter exhibited excellent photocatalytic degradation of methylene blue. The RGO/TiO_2 filter maintained its ability to degrade MB efficiently, even after five cycles of photocatalysis.

Keywords: Adsorption; Graphene; Photocatalysis; Titanium oxide; Water filter; Zinc oxide

1. Introduction

Impeccable hygiene is needed to maintain the high quality and shelf life of end-products. Purification of water is required to ensure safety and to achieve hygienic conditions. During the preparation, processing, manufacturing, and packaging of pharmaceutical and cosmetic products, foods and beverages, there is a need to maintain a high level of cleanliness and the purity of ingredients/raw materials to guarantee high standards and shelf life. Water is used in many of the requisite processes and so controlling its purity and removing pathogenic organisms responsible for waterborne diseases such as cholera, typhoid fever, and dysentery (disinfection) is absolutely essential [1]. Most municipal water supply systems achieve disinfection by chlorinating the water because this is a highly efficient and cost-effective method. Filtration methods such as ultrafiltration, nanofiltration, microfiltration, and reverse osmosis can also be used when there is a need to remove organic contaminants like proteins, gelatins, and colour pigments, but in general this requires the use of state-of-the-art technologies and makes the process more expensive. There are other disinfectant methods of microbiological impurities such as ultraviolet (UV) and ozone plants. They have the advantage of not adding any other compound to the water but are not so effective in terms of purifying the water.

The current water purification strategies mentioned above have several drawbacks. In chlorination, chlorine, chlorine oxide, or orhydrochlorous acid is added to the water to kill micro-organisms. These chlorine compounds can also react with naturally occurring organics, however, resulting in the formation of carcinogenic halo-organic compounds [2]. As a consequence, the quality of the water needs to be monitored [3]. In filtered systems, it is necessary to replace the filters frequently in order to meet the demand for high-quality water. Scheduled replacement of the filters increases the system's downtime and hence the operating costs. Conventional

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technologies, including sedimentation, filtration, chemical, and biological methods, merely transform the contaminants from the liquid to the solid phase, thus creating secondary pollutants [4]. UV disinfection of water requires the use of high-energy photons which are hazardous to health and expensive because of the large amounts of electric power needed to generate radiation. Ozone treatment of water requires the ozone to be manufactured on site and requires special handling because of its unstable nature and tendency to decompose with explosive violence.

In view of the disadvantages of the existing water treatment technologies, advanced oxidation processes (AOPs), which employ a combination of heterogeneous catalysis and radiation, are becoming more important in wastewater treatment. The main advantage of AOPs compared with other technologies is their capacity to remove organic contaminants from the environment in both the gaseous and aqueous phases by converting them into the simplest organic compounds and eventually mineralising them to harmless elements like carbon dioxide and water [5]. The AOPs comprise Fenton reaction (OH radicals), photo-Fenton, ozonation, ultrasonically assisted catalysis (sonocatalysis), photon-assisted catalysis (photocatalysis), and photo-electro-catalysis whereby the photon-absorbing material is separate from the catalyst. Among AOPs, heterogeneous photocatalysis and photoelectrocatalysis, which employ semiconductor catalysts such as TiO_2 and ZnO, have a proven record of efficiently degrading a wide range of refractory organics into biodegradable compounds [6].

The photodegradation process is independent of the state (gas or liquid) of the contaminant and so it is a viable decontamination process with a significant number of applications [7,8]; as a consequence, interest in this area has grown since the 2000s. Its technological applications, however, seem to be limited by the need to use an ultraviolet (UV) excitation source to initiate the chemical process. If the photodegradation process could be changed so that the chemical processes causing the degradation of the contaminants could be initiated with solar light, there would be significant energy and cost-saving benefits. Moreover, the water decontamination processes could be implemented where they are needed most in developing and third-world countries where clean water and abundance of electrical energy are generally not available. Therefore, the development of a photodegradation catalyst which uses solar light is an appealing challenge. In photocatalytic degradation of organic contaminants, the process starts with the absorption of a photon by the semiconductor, resulting in the promotion of an electron from the full valance band into the empty conduction band, thus creating an electron–hole pair. Both the electron and hole can then migrate to the surface of the semiconductor where there is electron transfer between the organic contaminants, water molecules and oxygen, creating both free radicals and charged species which react with each other causing the organic contaminant to decompose [9].

Nanocrystalline titanium dioxide (TiO_2), is widely used as a photoactive semiconductor for photoinduced processes and is a promising candidate for solar energy conversion applications such as photocatalysis, photochromism, and photovoltaics; its electrical properties depend on the morphology and shape of the nanocrystals [10]. In addition, the efficient electron transfer from the surface of the TiO_2 to oxygen in either water or an oxygen-rich environment means that irradiated TiO_2 has the ability to decompose and/or oxidise most organic and/or inorganic compounds [11,12]. Consequently, TiO_2 can be used in wastewater processing since organic contamination can be totally degraded and mineralised to CO_2, H_2O, and harmless inorganic anions. In addition, in a marine environment, the photocatalytic action of TiO_2 prevents the attachment of marine life (barnacles) to ships (antifouling agents) [13]. Zinc oxide is a well-known semiconductor and has a significant number of applications such as gas sensing [14], catalysis [15] and wastewater treatment [16]. Both TiO_2 and ZnO, however, have wide band-gap energy of 3.2 eV and 3.37 eV respectively, and so they are only active under near-ultraviolet irradiation. Thus, band gap engineering is required to shift the absorption range to the visible region. To achieve this shift for TiO_2 doping with non-metal ionic dopant series like carbon, nitrogen, boron and fluorine [17] is required. Furthermore, the presence of non-metal species also minimises the photogenerated electron–hole recombination rate, and thus improves quantum efficiency.

Graphite and graphene are very similar as both consist of hexagonal arranged carbon. The difference lies in the number of layers in the crystal. Graphite is a traditional crystal whereas graphene consists of only one or two layers. The physical arrangement of the atoms in graphene constrains the electrons to move in two dimensions. In addition, graphene is a semi-metal so there is a small overlap between the valance and conduction bands. With the addition of a gate electrode, electron and hole concentrations in the channel can be as high as 1013 cm^{-3} with mobility at room temperature in the order of 10,000 $cm^2\,V^{-1}\,s^{-1}$. These properties make graphene an ideal candidate for the next generation of electronic devices [18]. In addition, graphene has extremely good mechanical and thermal properties and thus has potential application in a wide variety of uses such as composite materials, fuel cells, batteries, chemical detectors, and solar cells [19–29]. Moreover, the integration of graphene with inorganic nanoparticles allows the properties of the nanocomposite to be engineered for specific applications [23,30–36] and is emerging as a class of new and exciting material [37].

The aim of the present work was to create a new type of water filter. A standard polypropylene (PP) filter was incorporated with reduced graphene oxide (RGO) and metal oxides via a simple hydrothermal approach. To test the modified filter, the photodegradation of methylene blue was investigated under the illumination of halogen light.

2. Experimental

2.1. Materials

The chemicals used in this research were graphite 3061 and titanium isopropoxide (99%) purchased from Asbury Graphite Mills Inc. (Asbury, NJ) and Acros Organic (Geel, Belgium),