



Superhydrophilic and self-cleaning rGO-TiO₂ composite coatings for indoor and outdoor photovoltaic applications



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ABSTRACT

The rGO-TiO₂ composite catalysts were successfully synthesized by adopting a simple solvothermal method followed by heating at mild condition of 300 °C for 1 h, in which graphene oxide (GO) was converted to reduced graphene oxide (rGO). Spin coating technique was used for coating TiO₂ and rGO-TiO₂ composite catalysts on Indium tin oxide (ITO) coated glass resulting in high transparency of about 75%. The rGO content was varied to optimize the activity of the catalysts. These synthesized catalysts were characterized by various techniques such as X-ray diffraction, Raman, Fourier transformed infrared and UV–vis diffuse reflectance spectral techniques, atomic force microscopy, transmission electron microscopy, scanning transmission electron microscopy and X-ray photoelectron spectroscopy. The anatase phase of TiO₂ was successfully achieved with crystallite size of less than 6.7 nm. Incorporation of spherical TiO₂ particles on rGO resulted in reduction of band gap from 3.05 to 2.74 eV. The interaction of TiO₂ with rGO sheet decreased the rate of electron-hole pair recombination and also facilitated the efficient transfer of electrons from TiO₂ to rGO. 0.5 wt% rGO loaded TiO₂ composite coatings showed highly efficient superhydrophilicity and self-cleaning property of decontaminating the adsorbed pollutants under simulated solar light irradiation within 30 min. A threefold higher photocurrent was generated by rGO-TiO₂ composite catalyst coating than pristine TiO₂ coating. This work affords a new pathway for efficient utilization of solar energy by using our synthesized catalysts for indoor as well as outdoor applications.

1. Introduction

Nanoscaled TiO₂ photocatalysis has attracted much attention due to its low cost, good stability, favourable chemical and physical properties and high photoactivity [1]. It has different commercial applications from indoor to outdoor constructions because of its unique properties [2]. The TiO₂ nanoparticles coated on glass surfaces were effectively utilized for self-cleaning application in various places such as National Opera Hall, China [3], Dives in Misericordia Church in Rome [4] and Roof of Dubai sports city's cricket stadium [5]. A surface coated with such material will reduce the maintenance cost of the construction by its self-cleaning property. The TiO₂ coated film shows water contact angle (WCA) of 72° in the absence of light. However upon irradiation with ultraviolet (UV) light, the WCA reduces to 0° which results in spreading of water droplets on the surfaces which is known as photo-induced superhydrophilicity and was first reported by Wang et al. [6]. The photocatalytic self-cleaning efficiency of the coating materials was investigated by its photo-induced superhydrophilicity and also the ability to decompose the adsorbed dirt or other pollutants [2]. The

photo-induced superhydrophilicity of TiO₂ involves different mechanism such as light-induced oxygen vacancy generation [6], reconstruction of surface hydroxyl groups [7] and removal of the carbonaceous layer present on surfaces of TiO₂ exposed to air [8]. From the invention of photocatalytic water splitting by Fujishima and Honda in 1972 [1], TiO₂ has been considered as an excellent photoactive material in photovoltaic devices and hydrogen production applications.

However, the fast recombination of electron-hole (e⁻/h⁺) pairs, lack of visible light absorption and low electrical conductivity (~1 × 10⁻¹² to ~1 × 10⁻⁷ S cm⁻¹) of TiO₂ become obstacles on its efficient utilization [9]. To overcome these issues, several efforts have been taken such as noble metal deposition, cationic and anionic doping, sensitization and addition of sacrificial agents [10–12]. Though the noble metal deposition has several advantages over others, high cost and low abundance of the noble metals restrict their use in large scale applications. Graphene is an appropriate replacement to noble metals since it has high surface area, high electrical conductivity (10⁶ S cm⁻¹), high carrier mobility (200,000 cm² V⁻¹ s⁻¹) and efficient electron (e⁻) transfer from TiO₂ to graphene as its redox potential is just below the conduction band (CB)

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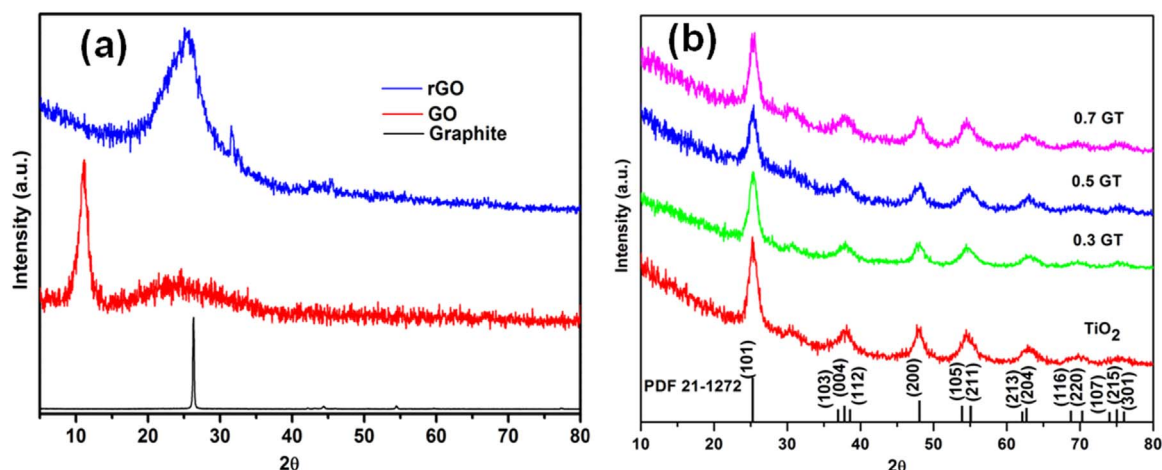


Fig. 1. XRD patterns of (a) graphite, GO and rGO and (b) TiO₂ and rGO-TiO₂ composites.

Table 1
Crystallite size and band gap of TiO₂ and rGO-TiO₂ composites.

Catalyst	Crystallite size (nm)	Band gap (eV)
TiO ₂	6.0	3.05
0.3GT	6.7	2.88
0.5GT	6.4	2.92
0.7GT	6.2	2.74

edge of TiO₂ [13]. There are several reports available on reduced graphene oxide (rGO)-TiO₂ pair having better performance [14–17]. The development of highly efficient rGO-TiO₂ materials are still needed for large scale applications because of the reasons such as weak interaction of rGO with TiO₂, insufficient active sites or surface area to interact with the surroundings, deadly synthetic procedure especially in reduction of graphene oxide (GO) and lack of cost-effective fabrication of devices. Therefore the urgent focus should be to address these issues.

In this study, efficient solar light active rGO-TiO₂ composite catalysts were synthesized by simple solvothermal method followed by heating at 300 °C. The anatase phase of TiO₂ on rGO with crystallite size less than 6.7 nm were obtained. The amount of GO was varied to optimize the activity of the catalysts. The rGO-TiO₂ composite coating with high transparency of about 75% was fabricated by spin coating technique. These synthesized catalysts were characterized by various techniques such as X-ray diffraction (XRD), Raman, Fourier transformed infrared (FT-IR) and UV–vis diffuse reflectance (UV–Vis–DRS) spectral techniques, atomic force microscopy (AFM), transmission

electron microscopy (TEM), scanning transmission electron microscopy (STEM) and X-ray photoelectron spectroscopy (XPS). The band gap of the composite catalysts reduced from 3.05 to 2.74 eV. The reduced rate of e⁻/h⁺ pair recombination and also the efficient transfer of electrons from TiO₂ to rGO were achieved by interaction of TiO₂ with rGO sheet. Under simulated solar light irradiation, the coating with 0.5 wt% rGO loaded TiO₂ composite (0.5GT) showed efficient superhydrophilicity, self-cleaning property and threefold higher photocurrent.

2. Experimental

2.1. Materials

Graphite flake, natural (10 mesh) was procured from Alfa Aesar, USA. Sodium nitrate (NaNO₃), sodium sulfate (Na₂SO₄), sulfuric acid (H₂SO₄), potassium permanganate (KMnO₄), hydrogen peroxide (H₂O₂, 30%), hydrochloric acid (HCl) and isopropyl alcohol (IPA) were procured from Merck Ltd., India. Titanium isopropoxide (TTIP) was procured from Aldrich, India. The microscopic glass slides were procured from Polar Industrial Co., India. ITO glass substrates (< 10 Ω, 1.1 mm thickness, > 90% transmittance) were procured from Shilpa Enterprises, India. All chemicals and reagents were of analytical grade and were used as such without further purification and Milli-Q (Millipore) water was used throughout this work.

2.2. Synthesis of GO and rGO

The GO was prepared by Hummers method with some modifications

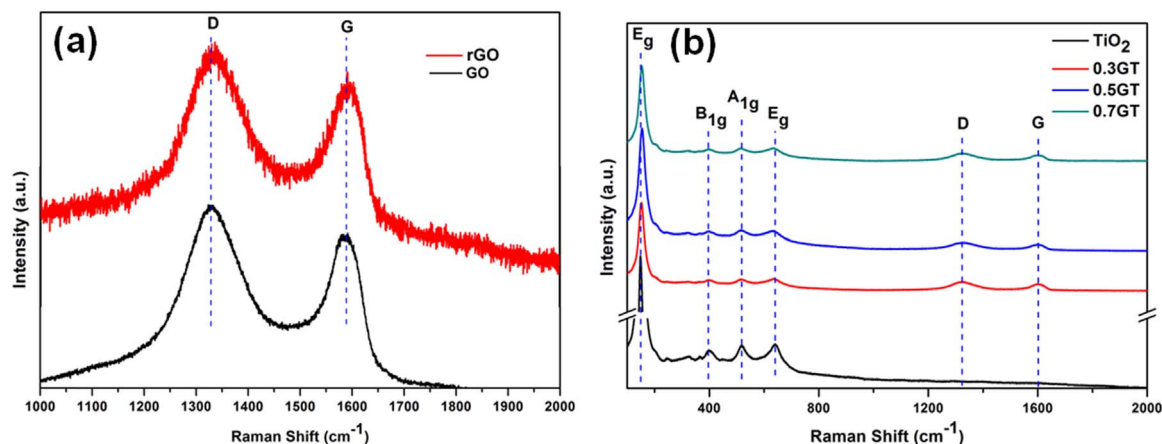


Fig. 2. Raman spectra of (a) GO and rGO, (b) TiO₂ and rGO-TiO₂ composites.

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