

Structural and photophysical studies of few layers of reduced graphene oxide functionalized with Sn(IV) tetrakis (4-pyridyl)porphyrin dichloride



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ABSTRACT

Reduced graphene oxide has attracted great interest owing to its exceptional properties, and its combination with Porphyrins has many potential applications in optoelectronics, light emitting diodes and dye-sensitized solar cells, because of the long excited-state lifetimes and delocalizable electron density on the porphyrin. Few layers of reduced graphene oxide (FRGO) were synthesized and functionalized with Sn(IV) tetrakis(4-pyridyl) porphyrin dichloride (SnTPyP²⁺). The structural and photophysical properties of the solution-cast thin films prepared from this composite were investigated by means of Scanning electron microscopy, X-ray diffraction, Raman scattering, FT-Infrared, UV–vis absorption, steady state Photoluminescence (PL), and time resolved PL (TR-PL). The results showed strong interactions between these hybrids components and successful deposition of the porphyrin on the FRGO layers. Moreover, a steady state PL quenching of the SnTPyP²⁺-FRGO composite film indicated an important interaction suggesting a photoinduced electron transfer between SnTPyP²⁺ and FRGO. The TR-PL measurements of the photogenerated charges lifetimes showed major change for the SnTPyP²⁺ after its interaction with the FRGO, resulting in a decrease in the PL mean lifetime.

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1. Introduction

Graphene is a two-dimensional (2-D) nanomaterial consisting of sp²-hybridized carbon atoms forming a one-atom thick honeycomb lattice. Owing to its unique structure and electronic properties, it has attracted the attention of scientists in recent years [1–3]. Graphene oxide (GO) sheets can be readily prepared by oxidizing graphite according to the Hummer's method [4]. The exfoliated graphene oxide sheets (GO) were suspended in polar solvents and reduced to produce reduced graphene oxide (RGO) [4,5]. The thermal, chemical, sonochemical, microwave, photocatalytic, or electrochemical treatment used to reduce GO can only partially restore aromatic sp² structure of graphene. By varying the

degree of GO reduction, it is possible to tune the optical properties of RGO [6,7].

Recently, there were a few papers reporting multiple-bilayered structures of RGO composite films that were hybridized with porphyrin derivatives using layer-by-layer method [8]. In the fabrication, the bilayered structures were constructed mainly based on the π–π and/or the electrostatic interactions between the RGO sheets and the other component. To date, the compounds used to construct multiple-bilayered structures with RGO include nanoparticles and polymers [9,10]. It is important to study the structural and optical properties of RGO with luminescent molecules such as porphyrins that play an important role when combined with electron donors or acceptors in mimicking light harvesting and photoinduced electron transfer, suggesting that porphyrin molecules could serve as potential photosensitizers in dye-sensitized solar cells as well as electron donors in bulk heterojunction solar cells applications [11,12].

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Another interesting aspect of the porphyrins is their ability to interact with various carbon nanomaterials, such as graphite and carbon nanotubes (CNTs), through π -stacking that takes place between their electron-aromatic cores and conjugated surface of the carbon nanomaterials [13–15]. Another interesting aspect to explore is the dispersion of few layers of RGO (FRGO) in Sn(IV) tetrakis(4-pyridyl) porphyrin dichloride (SnTPyP²⁺) aqueous solution. It is known that the photoexcited states lifetime of porphyrin is long enough to interact with the neighboring component so that electron and energy transfer become possible. It has been reported in our earlier work that interaction of FRGO and GO with nanostructured (self-assembled) porphyrins has been marked by a wide range of photoluminescence or fluorescence quenching depending on the pH and on the reduction level of GO [16–18].

In the present work, we prepared and studied the structural and emission properties of the FRGO covered with SnTPyP²⁺, for a profound understanding of mutual interaction between this porphyrin and FRGO. SEM, X-ray diffraction, Raman spectroscopy, Infrared spectroscopy, UV–vis absorption, steady state photoluminescence (PL) and time-resolved (TR-PL) were used in order to probe the structural and electronic properties of this donor/acceptor system for use in optoelectronic devices, sensors and particularly photovoltaic cells.

2. Experimental

2.1. FRGO/SnTPyP²⁺ composite preparation

The GO was synthesized from graphite using modified Hummer's method and the obtained solution was reduced with hydrazine hydrate. The obtained powder is dissolved in water and mixed using ultrasonication treatment for 45 min. More details of this preparation method of RGO were reported in our previous work [16,18]. The Sn(IV) tetrakis(4-pyridyl) porphyrin (SnTPyP²⁺) dichloride was purchased from Frontier and used as received. This molecule was combined with Meso-tetrakis (4-phenylsulfonic acid) porphyrin [H₄TPPS₄²⁻] as precursor for porphyrin nanorods structure already studied in our earlier work [16,18]. The investigation of the interaction of the positively charged porphyrin at different concentration with the FRGO is presented in this work. The solution of the FRGO (3 ml) was mixed with SnTPyP²⁺ (1 ml). Then, the prepared mixture was put into an ultrasound bath for 15 min at room temperature to ensure homogeneity. Finally, a thin

film of the FRGO-SnTPyP²⁺ was deposited by drop casting on a glass substrate which was already rinsed with distilled water and acetone, and dried in an oven for 10 min.

2.2. Characterisation

SEM images were obtained using a JEOL JSM 7600F, X-ray diffraction (XRD) patterns of FRGO, SnTPyP²⁺ and the composite were performed with Bruker's D8 advanced X-ray diffractometer using CuK α radiation ($\lambda = 1.5418^\circ \text{ \AA}$). Raman spectra were carried out using Jobin Yvon T64000 Raman spectrometer (gratings with 1800 grooves/mm) at excitation wavelength of 514 nm, laser power was adjusted to 7 mW. Infrared absorption measurements were performed using Fourier transform VERTEX 70 Series FTIR spectrometer of Bruker Rock Solid interferometer. UV–vis spectra were measured on a PerkinElmer LAMBDA950/1050 UV/VIS/NIR spectrophotometers are dual-beam, dual-monochromator instruments equipped with an optional 150 mm integrating sphere accessory.

Photoluminescence measurements in Steady-state were carried out at room temperature with a Jobin Yvon Fluorolog-3 spectrometer using a Xenon lamp (500 W) and a CCD detector with excitation wavelength of 420 nm. Time-resolved photoluminescence (TR-PL) experiments were performed with a regenerative amplified femtosecond Ti:Sapphire laser system (Spectra Physics Hurricane X). This setup generates 100 fs pulses at 800 nm with a repetitive rate of 1 kHz and a nominal power of 1 W. The laser line is frequency-doubled with a thin BBO crystal to obtain an excitation $\lambda_{\text{exc}} = 400 \text{ nm}$ (3.1 eV). Emission spectra are temporally resolved with a high dynamic range Hamamatsu C7700 streak camera coupled to an imaging spectrograph with a temporal resolution less than 20 ps and processed using the HPDTA Hamamatsu software.

3. Results and discussion

3.1. Scanning electron microscopy (SEM)

The main objective of our study is the inclusion of RGO in a porphyrin in order to create an interaction with improved charge transport properties. Fig. 1 shows the images recorded by scanning electron microscopy (SEM) of RGO, SnTPyP²⁺ and RGO-SnTPyP²⁺ composite films. The RGO image in Fig. 1(a) shows large

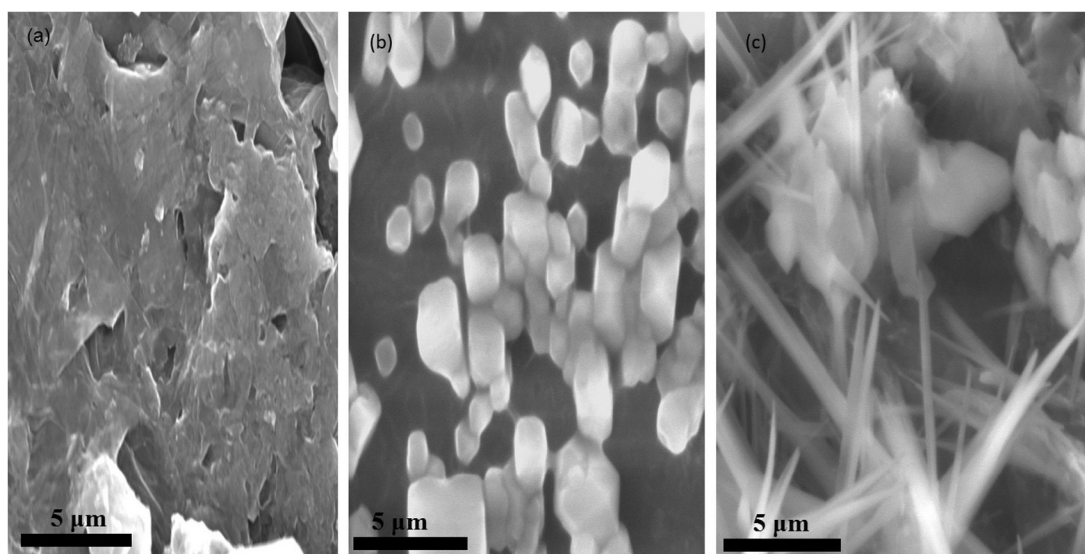


Fig. 1. SEM images of FRGO (a), SnTPyP²⁺ (b) and FRGO-SnTPyP²⁺ (c), acquired at 20 kV.

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