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Photoconductivity enhancement of reduced graphene oxide with reduced oxide graphene quantum dots hybrids film

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ABSTRACT

A hybrids film for reduced graphene oxide with reduced graphene oxide quantum dots (rGO+rGOQDs) was simply prepared by irradiation in Ar atmosphere. The rGO+rGOQDs film has better photoconductivity properties compared to the reduced graphene oxide (rGO) film. The photocurrent responsivity and external quantum efficiency values for the rGO+rGOQDs film reach ~120 mA W^{-1} and ~34% at 1 V, respectively. The photoconductivity enhancement is attributed to the strong absorption of rGOQDs and the photogenerated charge transfering from rGOQDs to rGO film.

1. Introduction

Graphene is a promising material for optoelectronic devices due to its high carrier mobility, and photon absorption [1,2]. However, the bandgapless for intrinsic graphene impedes its application in optoelectronic devices. Bandgaps of reduced graphene oxide (rGO) can be tuned by modifying the groups [3], which have been extensively investigated as the substitute for intrinsic graphene. Therefore, the rGO shares intriguing optoelectronic properties to offer various applications [4,5]. Unfortunately, the photocurrent responsivity (R) and external quantum efficiency (EQE) for rGO film were still low from our experimental results [6] and the previous reports [4].

Graphene quantum dots (GQDs) hold the charge storage in the trapping levels under the exciton confinement and quantum size effect [7,8]. Photodetectors based on the GQDs exhibit excellent stability and reproducibility, fast response speed, and high durability. Zhang [9] reported a high-performance deep-ultraviolet photodetector based on GQDs fabricated via a facile solution process. Ye [10] synthesized the GQDs that emit light from blue-green (2.9 eV) to orange-red (2.05 eV) by changing size, functionalities and defects. Although GQDs has good optoelectronic properties, its carriers can not be effectively transported to electrodes due to the poorly coupling with each other [11].

Therefore, a hybrid structure is designed by integrating the excellent optical and electrical properties of GQDs and rGO film to generate unique optoelectronic performances. We report the photoconductivity of the hybrids films for reduced graphene oxide with reduced graphene oxide quantum dots (rGO+rGOQDs) by photochemical reduction [6]. The results are shown that rGO+rGOQDs hybrids film has obvious photoconductivity enhancement compared to rGO film.

2. Experimental procedure

GO sheets were synthesized by modified Hummers' method [6]. Graphene oxide quantum dots (GOQDs) were fabricated using following procedure [12,13]. 1.6*g* XC-72 carbon black was put into 300 mL 15 mol/L nitric acid. Then, the mixture was heated at 160 °C for 24 h. After heated at 180 °C to evaporate water and concentrated nitric acid, a light black solid was obtained. Then, the dried sample was dissolved in approximately 800 mL deionized water, and centrifuged at 12000 rpm for 30 min to obtain a supernatant. Finally, the supernatant was filtered through 220 nm and 25 nm microporous membrane in vacuum, and then the brown GOQDs were obtained. The GO suspension (1 mg/mL) in ethanol was spun on pre-cleaned SiO_2 substrate to prepare GO films. Then, the GOQDs suspension (0.5 mg/mL) in ethanol was spun on the GO film. Finally, the hybrids films were sealed in quartz tube and irradiated with ultraviolet light from a 500 W high-pressure Hg lamp in pure Ar atmosphere for 60 min, and the rGO +rGOQDs hybrids films were obtained.

The morphologies and microstructures of the samples were in-

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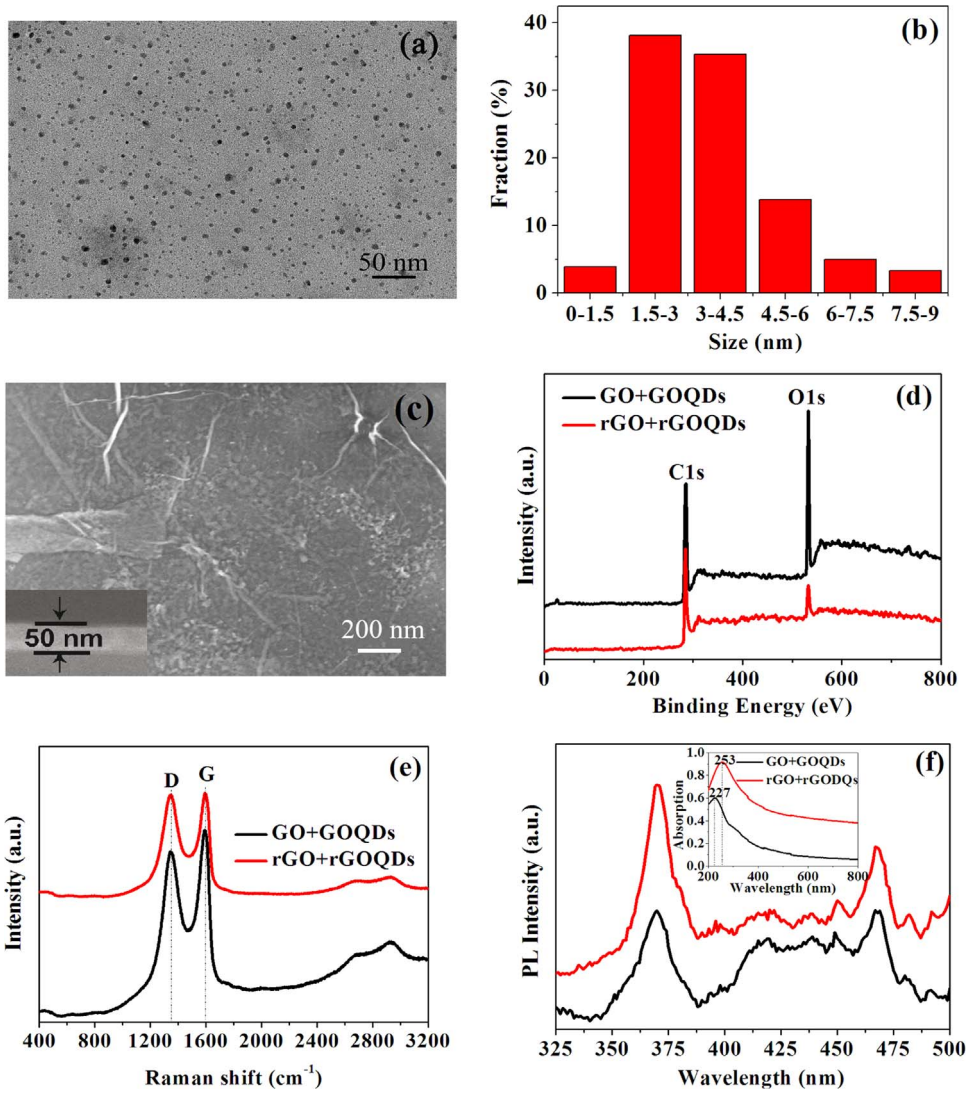

Fig. 1. (a) TEM image of GOQDs. (b) Diameter distribution of GOQDs. (c) SEM image of the rGO+rGOQDs film. (d) XPS spectra of GO+GOQDs and rGO+rGOQDs films. (e) Raman spectra of GO+GOQDs and rGO+rGOQDs films. (f) PL spectra of GO+GOQDs and rGO+rGOQDs films excited at 260 nm wavelength (Insert is UV–vis–NIR absorption spectra).

vestigated by transmission electron microscopy (TEM, JEM-2100), field emission scanning electron microscopy (FE-SEM, Hitachi S-4800). The X-ray photoemission spectroscopy (XPS) measurements were performed on PHI5000 Versa Probe (ULVAC-PHI) using Al Ka radiation. Optoelectronic characteristics were measured by a four-probe method under ambient condition [6].

3. Results and discussion

Fig. 1a and b show the TEM image and diameter distribution of GOQDs. The GOQDs are relatively uniform with diameters of approximately 1.5–5 nm, and the average diameter of GOQDs is ca. 2.5 nm, which is smaller than the reported 3.0 nm [12]. Fig. 1c is the representative SEM image with a few wrinkles and the thickness of ~50 nm for the rGO+rGOQDs film. Fig. 1d reveals the C and O contents obtained from XPS measurements of the rGO + rGOQDs and GO +GOQDs film. The rGO+rGOQDs film has a relatively low O content defined as 100 O/C at% (ca. 18.9 at%) compared to GO+GOQDs film (ca. 46.4 at%). It suggests that the majority of oxygen groups in rGO +rGOQDs film are removed after irradiation, which is similar to our previous work [6]. The XPS results of the GOQDs and rGO films (not shown here) are same to reported results for Sun [13] and Li [14]. Fig. 1e shows difference of Raman spectra between the rGO+rGOQDs and GO+GOQDs films. There is no obvious frequency shift of the G-band and D-band for two films. The D/G intensity ratios (I_D/I_G) for the GO+GOQDs and rGO+rGOQDs films are about 0.90 and 0.99, respectively, which indicates that the rGO+rGOQDs films have more defects and disorders than the GO+GOQDs film [15].

Fig. 1f indicates the PL spectra of the two films with an excitation wavelength of 260 nm. There are two obvious emission peaks and some weak emission peaks for GO+GOQDs and rGO+rGOQDs films in range from wavelength 320–500 nm. Compared to GO+GOQDs film, rGO +rGOQDs film exhibits clear PL enhancement. It derives from the functionalized groups and some oxidized sp^3 sites, such as C-O, C=O and O=C-OH, have important contribution to the fluorescence [16]. The UV–vis–NIR spectra of GO+GOQDs and rGO+rGOQDs films are shown in Fig. 1f (Inset). The UV–vis–NIR light absorption of rGO +rGOQDs film resulted in significant increase compared to that of GO +GOQDs film. The bathochromic shift (~27 nm) of the absorption maximum of rGO+rGOQDs film compared to that of GO+GOQDs film, which also suggests an extended π-stacking interlayer conjugation arising from GO+GOQDs to rGO+rGOQDs film [15].

Fig. 2a shows current-voltage curve of the rGO+rGOQDs film (GO +GOQDs film shows no photoresponse effect due to its insulation to some extent) with/without power 60 mW white-light illumination, and the photocurrent is very obvious. The photocurrents of the rGO

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