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Continuous gas-phase synthesis of graphene nanoflakes hybridized by gold nanocrystals for efficient water purification and gene transfection



Jeong Hoon Byeon a, Young-Woo Kim b,*

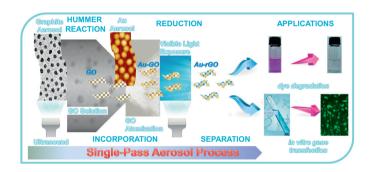
- ^a Department of Chemistry, Purdue University, IN 47907, United States
- ^b Department of Automotive Engineering, Hoseo University, Asan 336-795, Republic of Korea

HIGHLIGHTS

- A continuous gas-phase synthesis of gold-reduced graphene oxide (rGO) hybrid nanoflakes was employed.
- Spark discharge produced gold nanocrystals were deposited on rGO nanoflakes during atomization process.
- The ability to achieve dye degradation and gene transfection was greater than that of commercial materials.

G R A P H I C A L A B S T R A C T

Continuous gas-phase hybridization was employed to prepare gold-reduced graphene oxide hybrid nanoflakes as potential materials for efficient water purification and gene transfection.



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ABSTRACT

The present work reports for the first time a preparation of reduced graphene oxide (rGO) via a gas-phase process where a visible light photocatalytic reduction of GO with the use of gold (Au) nanocrystals was performed in a single-pass configuration. Primary Au crystals (\sim 4 nm in diameter) were quantitatively incorporated with GO (\sim 36 nm in lateral dimension) in the form of Au–GO hybrid nanoflakes (\sim 37 nm in lateral dimension). The hybrid flakes were then photocatalytically reduced into a form of Au–rGO, and thus the size and structure of the hybrid flakes were reduced (\sim 32 nm in lateral dimension) and changed (the ratio between the D and G bands increased from 0.82–1.10). The Au–rGO hybrid flakes were finally employed to degrade dye and transfect into cells $in\ vitro$, and the ability to achieve dye degradation and gene transfection was greater than that of commercial materials.

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1. Introduction

Graphene has attracted much interest for its unique physical and chemical properties and wide-ranging technological applications [1–3]. In the preparation of graphene, the method of utilizing graphene oxide (GO) as a precursor has been widely used [2]. GO is

an oxidized form of graphene with phenol hydroxyl and epoxide groups on the basal plane and carboxylic groups at the edges, and normally produced through processing graphite under oxidative conditions. Meanwhile, the reduction of GO has been widely used to prepare graphene [more accurately called reduced GO (rGO)], by methods such as chemical, thermal, flash, laser, or electrochemical reduction [1]. Among these methods, chemical reduction is considered to be the most effective and economical way to prepare rGO from GO. However, chemical reduction usually

^{*} Corresponding author. Tel.: +82 41 540 5819; fax: +82 41 540 5818. E-mail address: ywkim@hoseo.edu (Y.-W. Kim).

requires toxic chemicals, several tedious batch steps, high temperatures and energies, and special instruments and controls for the preparation of rGO, which limits its practical applications [1,4].

Photocatalytic reduction of GO has recently been proven to be an effective method to produce rGO [1]. Compared with conventional chemical reduction, the photoreduction of GO is green and easy to control *via* UV irradiation [5,6], and it often requires semiconductor nanoparticles with large band gaps such as TiO₂ and ZnO, used as photocatalysts, to accelerate the reduction. More recently, the surface plasmon resonance effect of noble metal nanoparticles on photocatalysis has attracted renewed interest, and it has been well established that GO with noble metal nanoparticles can be used as highly active photocatalysts in the presence of an electron donor [1,7,8].

Interest in nanocomposites or hybrid nanomaterials has been ever-growing, ascribed to their peculiarities in combining the desirable properties of building blocks for a given application [9]. Besides the applications of GO and rGO, it is a great desire to fabricate composites or hybrid materials which integrate GO or rGO with nanoparticles, polymers, or even nanotubes and fullerenes [10]. It is of great importance to bind metal nanoparticles onto GO or rGO because the combination and interaction between nanoparticles and GO or rGO will lead to multifunctional or even completely new properties in such a nanocomposite [11]. Metal nanoparticles incorporating GO were recently introduced as materials for antimicrobial [12], biosensor [13], and energy applications [14]. However, the metal hybridization of GO or rGO also requires additional chemical steps and controls for the preparation of metal nanoparticles [9,15], and thus it is still a challenge to prepare metal-(r)GO hybrid particles from raw graphite under continuous conditions.

The present work introduces continuous gas-phase approach for synthesizing gold (Au) decorated rGO hybrid nanoflakes and their applications for dye degradation and gene transfection in vitro. Freshly spark produced graphite nanoparticles were first immersed in an ultrasound-impinging device containing a simplified Hummer solution to form GO [4]. The reacted solution containing GO was injected into the reservoir of a collison atomizer. Another spark discharge generated Au nanocrystals, and the particle-laden flow passed over the collison atomizer orifice where they mixed with the atomized GO solution to form hybrid droplets. The droplets then passed through a denuder to drive solvent from the droplets, resulting in AuGO hybrid nanoflakes. The hybrid flakes were then immersed in another ultrasound-impinging device filled with ethanol to photocatalytically form Au-rGO hybrid nanoflakes under visible light. The hybrid nanoflakes were separated using a microfiltration kit, and finally they were applied to dye photodegradation and in vitro gene transfection measurements in mammalian cells. An electrical discharge as a graphite supplier was recently employed to prepare graphene nanoflakes [16], while a gas-phase process was also introduced to fabricate graphene with a unique structure [17]. These studies were the motivation to prepare Au–rGO by continuous gas-phase approach, and in addition there was no study to fabricate metal–rGO hybrid nanoflakes in a green and sustainable manner, although it has potential for a broad range of practical applications.

2. Experimental

Graphite nanoparticles were produced via spark discharge [18], and carried by nitrogen gas (99.9999% purity, 3 L min⁻¹) to an impinging device, as shown in Fig. 1. The specifications of the discharge configuration were as follows: electrode diameter (C-072561, Nilaco, Japan) and length, 3 mm and 100 mm, respectively; resistance, 0.5 M Ω ; capacitance, 1.0 nF; loading current, 2.0 mA; applied voltage, 3.0 kV; and frequency, 667 Hz. The impinging device, which contained a simplified Hummer's solution [4] and an ultrasound probe, was used to collect the graphite particles into the solution and react subsequently with the solution to form GO. The graphite particles experienced ultrasound (250 W cm⁻² in intensity) when they reached the gas (the graphite particle laden flow)-liquid (the simplified Hummer's solution in the impinging device) interface. GO was obtained by oxidation of the graphite particles (which acted as precursors) with 40 mL of H₂SO₄ and 1.8 g of KMnO₄. The residence time of the graphite particles in the impinging device was 3.8 min to form GO. The solution containing GO was injected into the reservoir of a collison atomizer using a peristaltic pump (323Du/MC4, Watson-Marlow Bredel Pump, US).

Another spark discharge between gold rods (AU-172561, Nilaco, Japan) was used to produce Au nanocrystals, and the particle laden flow was employed as the operating gas for atomizing the GO solution supplied by the pump. The Au crystals passed over the atomizer orifice, where they mixed with atomized GO droplets to form hybrid droplets. The droplets then passed through a denuder containing activated carbon pellets and silica gels to drive solvent from the droplets. The resulting aerosol Au–GO hybrid flakes were then injected into another impinging device containing ethanol with an ultrasound probe under visible light to photocatalytically form Au–rGO.

When Au nanocrystals are attached onto GO, a quasi-fermi level forms in the Au–GO flakes. The Au nanocrystals absorb visible light intensely because of the surface plasmon resonance effect, leading to an enhancement of the local electromagnetic fields near the rough Au surface by photoexcited metallic electrons and holes [1]. In the presence of ethanol the holes are scavenged to produce ethoxy radicals, thus leaving the electrons to accumulate within the Au crystals. The accumulated electrons serve to interact with the GO in order to reduce certain functional groups (Eq. (1)).

$$\mbox{Au} + \mbox{hv} \rightarrow \mbox{Au}(\mbox{h} + \mbox{e}) \xrightarrow{\mbox{c_2}\mbox{H_5OH}} \mbox{Au}(\mbox{e}) + \mbox{c_2}\mbox{H_4OH} + \mbox{H^+}$$

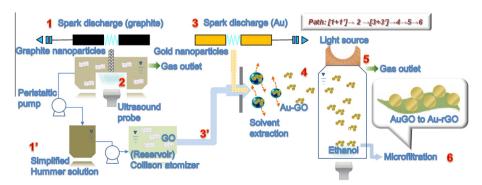


Fig. 1. Continuous gas-phase synthesis of Au-rGO using a serial reactor consisting of spark discharges, ultrasound probe inserted impingers, and a collison atomizer.

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