

## Review

## Graphene materials-based chemiluminescence for sensing



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## ABSTRACT

Graphene has attracted considerable attention in multidisciplinary research fields and shown various promising applications due to its unique structure and extraordinary physicochemical properties. This review covers the latest advances in graphene materials-based chemiluminescence (CL) for sensing. Chemiluminescence resonance energy transfer and luminescence quenching of graphene materials are discussed. Graphene materials, such as graphene nanosheets, graphene quantum dots, graphene oxide, and reduced graphene oxide have been employed successfully in CL systems in recent years. Graphene materials can be utilized as catalysts, platforms, and energy acceptors to improve the performance of CL. Possible challenges and future perspective on this topic are also presented.

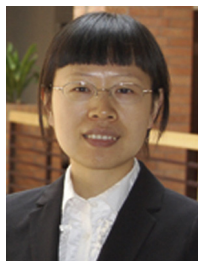
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## 1. Introduction

Chemiluminescence (CL) is a generation of electromagnetic radiation in the visible or near-infrared region. CL phenomena can take place in aqueous phases, on solid surfaces, and in gas phases, and are generated by redox reactions between at least two CL reagents under proper reaction conditions. During the CL reaction process, the produced electronically excited intermediate is unstable, and relaxes to the ground state with light emission [1]. CL is a powerful analytical technique with high sensitivity, wide linear range, simple instrumentation, and little background interference. It has been exploited in a wide range of applications throughout several different fields, including biotechnology, pharmacology, molecular biology, and environmental chemistry in the past few decades [2–4].

The primary disadvantage of CL analysis is its relatively weak luminescence due to the low quantum yield of inorganic molecules. Nanomaterials have been proposed to provide enhanced and amplified CL emission through catalytic process, energy transfer, surface plasmon resonance, or redox reactions [5–8]. Nanoparticle-assisted CL and its applications in analytical chemistry were explored in a particularly notable study by Giokas et al. [9]. Nanomaterial-based chemiluminescence resonance energy transfer (CRET) and its appli-

cations [10], as well as nanomaterial-amplified CL systems and their applications in bioassays [11] were researched at length in 2012. Lv et al. [12] summarized the analytical applications of nanomaterials in liquid-phase CL, including metal nanoparticles (NPs), magnetic NPs, quantum dots (QDs), and carbon-based nanomaterials. We ourselves published research on QDs-enhanced ultraweak CL systems, the mechanism of QDs in CL reaction, and their applications [13]. At this point, the most crucial research in this area focuses on identifying new nanomaterials with high CL efficiency to improve the sensitivity and stability of CL systems. Graphene materials show potential as effective CL reagents or as a novel alternative to catalyze redox CL reactions.

Graphene is a one-atom-thick 2D array of carbon atoms, packed in a honeycomb-shaped lattice with zero bandgap [14]. This  $sp^2$ -hybridized carbon material, has attracted extensive research interest in the fields of chemistry, physics, and material science since its first report in 2004 [15]. Favored for its extraordinary electron transfer rate, good quenching efficiency, large surface area and ultra-large planar structure, excellent carrier mobility and carrier capacity, and high mechanical properties, graphene has been widely successfully applied to transparent conductors, super capacitors, energy storage, and bio- and chemo- sensors in its relatively short history of research [16–18]. Graphene and its hybrids show many interesting optical properties due to their unique electronic structures. Despite the absence of bandgap, graphene still exhibits luminescence property through hot electrons [19]. Graphene QDs (GQDs), which have lateral dimensions less than 100 nm, show prominent edge effect and remarkably enhanced quantum confinement effect [20]. GQDs exhibit outstanding photoluminescence features, which is a phenomenon of light emission after the excitation of photons. The versatile photoluminescence properties of GQDs can be achieved by tailoring the intrinsic electronic structures of GQDs using size/shape controlling, or functional groups, chemical doping, or surfaces and edges modification. Compared with the fluorescence properties, GQDs can also be an emitting species in redox reactions generating strong chemiluminescence. During the chemical reaction, GQDs are directly oxidized to give rise to CL emission [21]. Graphene oxide (GO) is a partially oxidized graphene that involves functional groups of oxygen such as hydroxyl, carboxyl, and epoxy. These functional groups open up the bandgap of graphene in GO, endowing it with photoluminescence features. Fluorescence of GO, for example, displays special spectral properties with emission lifetime independent of wavelength [22]. Reduced graphene oxide (RGO) is obtained by reduction reaction of GO to lower its number of oxygen-containing groups. These oxygen-containing groups enable RGO to easily covalently conjugate with other molecules and improve RGO water solubility as compared to graphene nanosheets. But then, graphene nanosheets have broader absorption spectrum and higher electron transport property, thus higher luminescence quenching efficiency than RGO. The photoluminescence of RGO can be adjusted in a broad spectral range by manipulating the number of functional groups through reduction reaction [23]. The photoluminescence property makes graphene, GO, and RGO favorable potential candidates for optoelectronic and photonic devices. Graphene and GO are typically covered with metal NPs or hybridized with luminescent reagent or other catalysts for multipurpose applications.

Resonance energy transfer (RET) is a nonradiative (dipole-dipole) energy transfer process that occurs between a donor and an acceptor, such as fluorescence RET (FRET) and chemiluminescence RET [24]. The donors can be fluorophore, and chemiluminescent compound with acceptors as fluorophores (e.g., dyes, quantum dots), respectively. Amongst them, CRET occurs via the specific oxidation of a luminescent substrate that then excites a suitable acceptor molecule during chemiluminescence reaction without an

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