氏　　名	窦　相南（ドウ　シャンナン）
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学位論文題名	Study on Mechanism and Application of Novel Reactive Oxygen Chemiluminescence System Based on Nanoparticles (ナノ粒子による新規活性酸素・化学発光システムの発光メカニズムと応用に関する研究)
論文審査委員	主査　教　授　　内山　一美 委員　准教授　　中嶋　秀 委員　准教授　　加藤　俊吾 委員　(清華大学)　教授　　林　金明

【論文の内容の要旨】

Chemiluminescence is process when the electronically excited product of a chemical reaction relaxes to its ground state with emission of photons. CL have advantages for applications in analytical chemistry, such as high sensitivity, wide linear range, simple and inexpensive instrumentation, low background noise.

Reactive oxygen species (ROS) is a collective term of oxygen free radicals and molecule, which contain one or more unpaired electrons. The presence of unpaired electrons generally imparts a higher reactivity to the free radicals. ROS mainly include an excited state of oxygen molecules, ie, singlet oxygen molecules (1O2), three kinds of oxygen-containing free radicals, namely superoxide anion radical (.O2-), hydroxyl radical (.OH) and hydroperoxy radical (HO2.); nonradicals that are either oxidizing agents　that are easily converted into radicals, such as hydrogen peroxide (H2O2), lipid peroxide (ROOH), HOCl, ozone (O3), peroxynitrite (ONOO-).

Reactive oxygen species can generate electronically excited products, which emit the weak CL during their decay to the ground state. It is difficult to detect the light emission directly by CL techniques because of the ultra-weak CL emission from ROS. The emphasis of this article is on the development of novel reactive oxygen CL system based on nanoparticles. The mechanism and the potential application of the established CL system are investigated. The main contents in our work contains as follows:

(1) Firstly, chapter 1 introduced the concept of chemiluminescence and reactive oxygen species. The classification, generation and detection of ROS, the reactive oxygen chemiluminescence system including chemiluminescence from oxidation of lumiol by reactive oxygen speices and hydrogen peroxide- based chemiluminescence system are reviewed.

(2) Chapter 2 developed a novel synthetic method for fluorescent carbon nanoparticles (FCNs) using acetic acid (AC) as carbon precursor. The released heat was produced during the reaction between water and P2O5, which promote the carbonization reacton besides vaporizing the AC. It was noted that the FCNs exhibited λex-independent fluorescent feature, which was very different with those of previously reported carbon dots. The FCNs are the new bifunctional CL reagent molecule for H2O2-based chemiluminescence system because of its oxidation and sensitizer roles in the reaction with luminol, NaHCO3 and NaHSO3.

(3) Chapter 3 demonstrated strong chemiluminescence (CL) of nitrogen doped carbon dots (N-CDs) due to hydroxyl radical (•OH) induced electron-hole transition in N-CDs. The introduction of N-CDs improved the utilization of H2O2 and drastically enhanced the generation of •OH. A pre-mixed NCDs/H2O2 solution was utilized for selective quantification of Fe2+ in solution via CL-emission. The CL intensity of the system is dependent on the concentration of Fe2+. The N-CDs/H2O2 system enabled the detection of Fe2+ as low as 1×10-9 M with a linear dynamic range of 1.0×10-9-1.0×10-6 M. Significantly, no interference was observed when a mixed solution of Fe2+ and other cations such as Al3+, Fe3+, or Cr3+ were introduced to N-CDs/H2O2. The CL method have been applied to practical evaluation of N-CDs/H2O2 system for detection of Fe2+ in tap, lotus pond, and canal water samples.

(4) In chapter 4, Here we demonstrate the promising capability of molybdenum sulphide-quantum dots (MoS2-QDs) for generation of ROS, which leads to enhance chemiluminescence. We explored that hydroxyl radicals activate MoS2-QDs and generate active catalytic sites on its surface. The activated MoS2-QDs then generate ROS such as, hydroxyl radical (•OH), superoxide radical (•O2-) and singlet oxygen (1O2) in sufficient quantitiy. The MoS2-QDs provide a new pathway for ROS generation at whole pH-range, which effictively degrade the organic pollutants and can be use in chemo-dynamic theraphy.