Self-photo-oxidation for extending visible light absorption of carbon dots and oxidase-like activity

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一、簡述論文概要及重大發現

碳點(C-dots) 是一種新型的碳基納米材料，具有多種物理化學性質並且良好的生物相容性、獨特的光學性質、低成本、低毒性、體積小、並且對生態友善等多種優點，可應用於傳感器、信息加密、電池、催化、LED、光敏劑、醫學成像、藥物輸送。但大多數水溶性碳點是由紫外光激發並放出藍光螢光，這點大大的限制碳點在光動力療法(PDT) 的應用。故作者利用紫外光照射處理碳點以產生活性氧 (ROS)，所產生的 ROS 會導致碳點自氧化並增加體積，更可以擴展可見光吸收（400 nm-650 nm）並延長螢光波長而從藍色轉為綠色。此外，光處理碳點的催化效率是原始碳點的 11 倍。由於延長的激發光和可見的光氧化活性，光處理的碳點可作為 PDT 試劑，同時提供高效的癌症治療。這項研究提供了一種簡單的方法來修飾碳點，從而產生新的特性。

二、對論文內容的提問

由不同碳前驅物或透過不同合成方法製備的碳點會呈現不同的吸收波長並放出不同波長的螢光，而波長的長短會影響穿透組織的能力。想知道若用不同碳前驅物或不同方式所製備出來的碳點，是否一樣能使用紫外光修飾，來增加其功能。

三、論文的缺點與評論

本篇發現通過紫外光照射碳點所產生的 ROS 會導致碳點自氧化並增加體積，並擴展了碳點的應用。光誘導的自氧化闡明了碳點的自修飾特性，或許可以為未來相關研究提供啟示，例如，在碳點的光誘導自氧化過程中添加適當的分子或其他奈米材料可能有助於獲得具有設計特性的混合奈米材料，值得我們好好思考學習。

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A B S T R A C T

Carbon dots (C-dots) are attractive for their low cost, excellent optical properties, and low toxicity, but most C-dots are limited by their UV absorbance, especially for photodynamic therapy (PDT) applications. Developing rational and simple strategies to obtain C-dots with longer excitation/emission wavelengths remains a challenge. Previous work showed that C-dots can produce a high yield of reactive oxygen species (ROS) upon UV light illumination. We herein show that the produced ROS can lead to further oxidation and growth of C-dots, leading to visible light absorption (400 nm–650 nm) and extend fluorescence emission from blue to green. In addition, the light-treated C-dots showed boosted peroxidase and oxidase mimicking activity than the initial C-dots in the absence of light. In the presence of visible light, the catalytic efficiency of the light-treated C-dots is 11-fold much higher than the pristine C-dots. The light-treated C-dots can be used as a PDT agent due to the extended excitation and visible photo-oxidative activity, simultaneously providing a highly efficient cancer therapy. This study provides a simple way to modify C-dots leading to novel properties.

1. Introduction

Fluorescent carbon dots (C-dots) have drawn increasing attention over the last decade owing to their excellent optical properties, water-solubility, and biocompatibility [1–4], allowing a wide variety of applications including sensing, therapeutics, optoelectronics, and photocatalysis [5–9]. Most water-soluble C-dots absorb in the UV region and emit blue light. It would be useful to obtain C-dots with longer excitation and emission wavelengths. Besides, C-dots were rarely used as an oxidizing or reducing agent [10,11]. To improve the properties of C-dots, the main strategies are to use different starting materials [12–14], and perform post-synthesis modifications. For example, poly-(ethylene glycol)-coated C-dots showed brighter photoluminescence [15]. Se or Te-doped C-dots allowed a high antioxidation activity for scavenging free radicals [16,17]. Protein, lipid or cyclodextrin-modified C-dots showed versatile applications in biocatalysis [7,18–20]. However, these methods are often expensive and sometimes complex to perform.

Nanomaterials with enzyme-mimicking properties, also known as nanozymes, have attracted considerable attention due to their higher stability and lower cost than natural protein enzymes [21–24]. Light-activated nanozymes with the advantages of light-regulated activity and using molecular oxygen as a green oxidant were used in biosensing, pollutant removal, cleavage of DNA, and cancer photodynamic therapy (PDT) [25,26]. While PDT shows important benefits of minimal invasiveness, low toxicity, and widespread applicability [27–29], the UV excitation of blue-emitting of C-dots greatly limited their application in PDT. Our previous work showed that C-dots can produce reactive oxygen species (ROS) upon light illumination, which offered strong photo-oxidation activities [30,31]. The produced ROS may lead to further crosslinking and oxidation of C-dots, which may be a simple way to

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