



Contents lists available at ScienceDirect

Advanced Powder Technology

journal homepage: www.elsevier.com/locate/apt



Original Research Paper

Improved photodynamic efficiency for methylene blue from silica-methylene blue@tannic acid-Fe(III) ions complexes in aqueous solutions

Wenhao Wang, Lishuang Yang, Haibin Sun, Zanzhong Yang, Qingyang Du, Chengfeng Li*

School of Materials Science and Engineering, Shandong University of Technology, 255049 Shandong, PR China

ARTICLE INFO

Article history:
Received 15 August 2017
Received in revised form 8 November 2017
Accepted 11 November 2017
Available online xxxx

Keywords:
Photodynamic therapy
Singlet oxygen
Silica
Methylene blue
Tannic acid

ABSTRACT

To avoid multidrug resistance and tumour recurrence, photodynamic therapy (PDT) was emerging as an alternative therapy and its efficiency was related to photosensitizer (PS) efficiency, oxygen concentration and light characteristic. Methylene blue (MB) molecules as PSs were loaded in silica (silica-MB) and followed by encapsulation by coordination complexes of tannic acid (TA) and Fe(III) ions. In comparison with those of silica-MB, decreased condensation of Si-O-Si, shifted infrared absorbance frequencies of chemical bands, delayed thermal degradation and modulated release behavior of MB were observed for silica-MB@TA with a core-shell structure. Although MB dimers were dominantly released from silica-MB, release of MB monomers from silica-MB@TA was significantly promoted, which was described by the Higuchi model. The promotion of release of MB monomers from silica-MB@TA indicated the well control of aggregate states of MB by the encapsulation of TA and Fe(III) ions complexes. Through monitoring the oxidation of uric acid, generation efficiency of singlet oxygen (1O_2) by MB released from silica-MB@TA was fairly higher than that from silica-MB. A facile method to encapsulate silica-MB with complexes of TA and Fe(III) ions was herein demonstrated to raise the generation efficiency of singlet oxygen.

© 2017 Published by Elsevier B.V. on behalf of The Society of Powder Technology Japan. All rights reserved.

1. Introduction

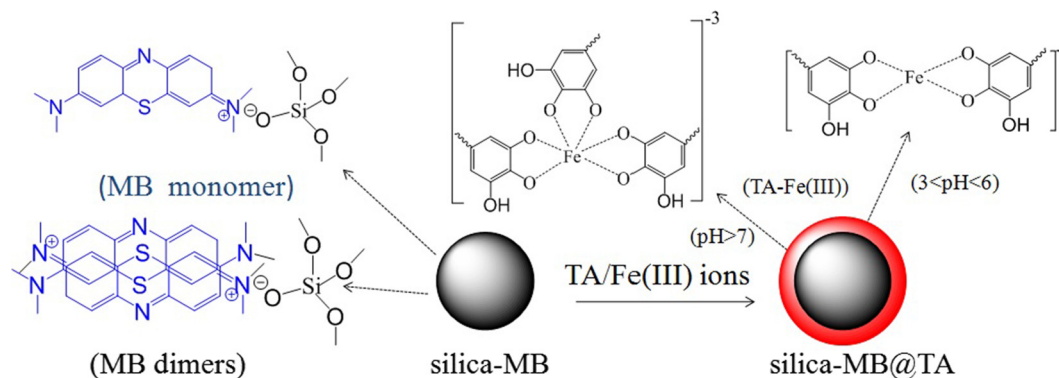
As a highly lethal disease, cancer is a serious threat to the health and life of human being. During cancer treatment by chemotherapy, subtherapeutic use and overuse of antimicrobials could cause the occurrence of multidrug resistance of microorganisms [1], which has become an obstacle to damage key macromolecules of tumour cells due to drug efflux from these pathological cells [2-4]. Photodynamic therapy (PDT) is emerging as an alternative therapy to damage proteins of tumour cells through necrotic and apoptotic pathways [5-7], and thus tumour drug resistance and the associated tumour recurrence were avoided [8,9]. The PDT efficiency was related to photosensitizer (PS) efficiency, light characteristics (intensity and wavelength) and oxygen concentration [10].

As a cost-efficiency PS, methylene blue (MB) has a high quantum yield of singlet oxygen (1O_2) generation ($\Phi_\Delta \sim 0.5$) and low dark toxicity [11,12]. However, raising the dosage of administrated MB would cause the formation of MB dimers, which are not only

generating 1O_2 in a less-effective way, but also quenching coexistent photoactive species [13,14]. Various nanoformulations have thus been demonstrated to load MB on surfaces of gold, silica, or carbon-based nanostructures [11,15-18] to increase the photostability of PS through controlling the aggregate state and avoiding self-quench effect of MB [19]. Stimuli-responsive release of loaded drugs from vehicles could be generally modulated through surface modification with grafting the disulfide bond-reducing molecules [20], pH-dependent supramolecular nanovalves [21,22] and protease-responsive cap system [23].

Although great efforts had been devoted to alter the release behavior of MB molecules, the aggregate state of released MB was only controlled in a limited way during outward diffusion of MB molecules from these vehicles with either encircling surfaces or unlocking pores. In our previous report, MB dimers were dominantly released from silica-MB@octacalcium phosphate powders with a network consisted of polyethylene glycol, citric acid, ethylenediaminetetraacetic acid and octacalcium phosphate on silica-MB [24]. Until now, promotion of releasing MB monomers from carriers were rarely reported. Herein, a modified Stöber method was firstly employed to synthesize MB-loaded silica (silica-MB), and then coordination complexes of tannic acid (TA)

* Corresponding author.
E-mail address: cfl@sdu.edu.cn (C. Li).



Scheme 1. Schematic illustration of synthesis process of silica-MB@TA.

and Fe(III) ions were assembled on the surface of silica-MB as shown in Scheme 1. Release behavior of MB monomers, degradation of drug carriers and generation efficiency of 1O_2 of released MB were finally investigated and discussed in detail.

2. Materials and methods

All chemical reagents with analysis purity were supplied by Sinopharm Chemical Reagent Co. Ltd. (China) and used without any further purification. All water was doubly-distilled and de-ionized.

2.1. Synthesis of silica-MB

A modified Stöber method was used to synthesize silica-MB in the following sequence [25,26]. Briefly, 92 ml of ethanol, 17.20 ml of water and 2.48 ml of $NH_3 \cdot H_2O$ were mixed and followed by addition of 0.10 g of MB under stirring. After 15 min, 3.44 ml of tetraethoxysilane (TEOS) was added and stirred for another 4 h. Silica-MB precipitations were collected by centrifuge and washed with ethanol twice. Colloidal solution of silica-MB was prepared in 20 ml of water. Silica-MB particles were collected by centrifuge of colloidal solution of silica-MB and dried at 70 °C for 24 h.

2.2. Synthesis of silica-MB@TA

Coordination complexes of TA and Fe(III) ions were used to coat silica-MB colloids by a one-step assembly method reported previously [27]. 0.30 ml of $FeCl_3 \cdot 6H_2O$ solution (Fe(III) ions solution, 10 mg/ml, the pH value of 2.80 adjusted by $NH_3 \cdot H_2O$) and then 0.30 ml of TA solutions (40 mg/ml) were added to aqueous silica-MB template suspension (5 ml). The suspension was vigorously mixed for 1 min immediately after the individual additions of Fe(III) ions and TA solution. The pH value of this suspension was subsequently raised by adding 100 ml of phosphate buffer saline (PBS, pH = 7.2–7.4). The particles were washed twice to remove excess TA and Fe(III) ions. In the washing step, the particles were collected by centrifugation and washed with ethanol and water twice. The remaining powders of silica-MB@TA1 were dried at 70 °C for 24 h.

Addition of Fe(III) ions (0.90 ml or 1.80 ml) and TA (equivalent volume of Fe(III) solution) solutions were altered from the standard conditions described above, while the other variables were kept constant. In this step, sample of silica-MB@TA3 or silica-MB@TA6 was synthesized.

2.3. Release behavior of MB

0.02 g of silica-MB and silica-MB@TA were incubated in 25 ml of PBS or acidic buffer (pH = 1) under rotary shaking (120 rpm) at 37 °C. The ultraviolet-visible spectroscopy (UV-Vis) absorption spectrum of the supernatant was recorded at different time intervals using a UV-Vis spectrophotometer (TU1901, Beijing, China). Different theoretical models were employed to simulate the release behavior of MB from carriers. The degradation percent was calculated according to the residual weight ratio after incubating 0.02 g of silica-MB and silica-MB@TA in 25 ml of PBS or acidic buffer (pH = 1) for 72 h. Silica-MB or silica-MB@TA was etched in the solution containing 26% of hydrofluoric acid, 50% of ethanol and 24% of deionized water to determine the loading capacity of MB molecules.

2.4. Evaluation of photodynamic activity

A solution of uric acid (3 mmol/l) was prepared through dissolving uric acid in a NaOH solution (2 mol/l) and then diluted to a

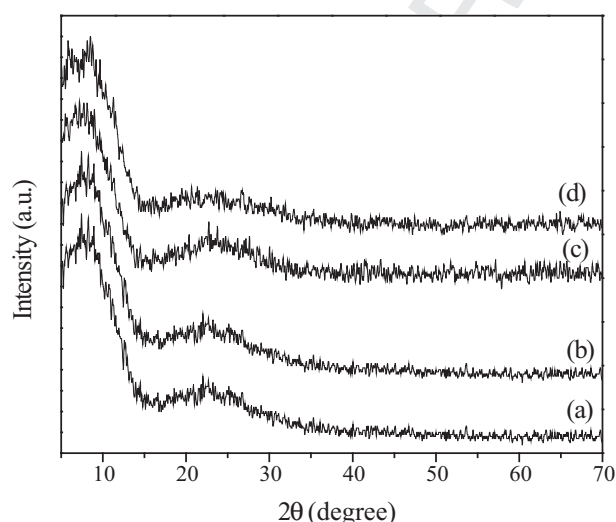


Fig. 1. XRD patterns of silica-MB (a), silica-MB@TA1 (b), silica-MB@TA3 (c), and silica-MB@TA6 (d).

Table 1
Element analysis by EDS of silica-MB and silica-MB@TA3.

Sample	Atomic percent (%)					
	C	N	O	Si	S	Fe
Silica-MB	29.80	4.25	53.32	12.25	0.38	0
Silica-MB@TA3	28.81	7.72	51.05	12.09	0.30	0.03

Download English Version:

<https://daneshyari.com/en/article/6577419>

Download Persian Version:

<https://daneshyari.com/article/6577419>

[Daneshyari.com](https://daneshyari.com)