



Synthesis of polypyrrole nanowire network with high adenosine triphosphate release efficiency

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ABSTRACT

A novel drug release system based on polypyrrole (PPy) nanowire network is developed for controlled adenosine triphosphate (ATP) release. Interestingly, the formation of the PPy nanowire networks is induced by ATP itself, i.e. ATP serves as both the morphology-directing agent and the model drug. More importantly, it should be pointed out that using ATP as morphology-directing agent for the formation of the PPy nanowire network can significantly increase the ATP release efficiency due to the high surface area of the resulting nanowire network. The experiment results show that ATP release efficiency increases from 53% (for conventional cauliflower-like PPy) to 90% (for PPy nanowire network) within 45 h upon electrical stimulation.

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1. Introduction

During the past decades, conducting polymer (CP) nanostructures have become a rapidly growing field of research because they usually exhibit novel properties based on their nanoscale size. PPy nanowire network is one of the most common nanostructures of CPs and has been synthesized by many research groups using various methods, such as functional molecule-induced synthesis [1–6], seeded growth [7], and interfacial polymerization [8]. The applications of the PPy nanowire network in the fields of biosensors, energy source, and electronics have also been extensively explored [9]. However, to the best of our knowledge, there is no report on the use of PPy nanowire network for controlled drug release. Considering the widespread application of conventional PPy in this field [10], it is necessary and important to investigate the drug release function of the PPy nanowire network.

In the present work, a drug release system based on PPy nanowire network is developed for controlled adenosine triphosphate (ATP) release. Interestingly, the formation of the PPy nanowire network is induced by ATP itself, i.e. ATP serves as both the morphology-directing agent and the model drug. ATP is recognized as an important neurotransmitter or co-transmitter in both the central and peripheral nervous system, and thus its incorporation into a CP matrix could be of great interest in the study

of cell metabolism, neuron science, and drug delivery [11]. As a result, ATP has become one of the most frequently reported model drugs related to CP-based drug delivery systems [12–18]. However, so far it has not found application as morphology-directing agent in the synthesis of CP nanostructures. It should be noted that using ATP to guide the growth of the PPy nanowire network for controlled ATP release could significantly increase the ATP release efficiency due to the high specific area of the nanowire network. Our experiment results show that ATP release efficiency increases from 53% (for conventional cauliflower-like PPy) to 90% (for PPy nanowire network) within 45 h upon electrical stimulation. Furthermore, self-powered ATP release from the PPy nanowire network is achieved by coating a thin magnesium layer on the surface of the nanowire network. It is found that ATP release efficiency of the self-powered system is also much higher than that of the non-nanostructured PPy (coated by magnesium layer as well) and the ATP release rate can be tuned by forming a patterned magnesium layer.

2. Experimental

2.1. Chemicals

Pyrrole was purchased from Sinopharm Chemical Reagent Co., Ltd. (China), distilled under the protection of nitrogen gas and stored frozen. ATP was purchased from Sigma. All other chemicals were of analytical grade and were used as received.

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2.2. Synthesis of PPy nanowire network

The PPy nanowire network was synthesized electrochemically at room temperature in a one-compartment cell by the use of LK2005A Electrochemical Workstation (Lanlike Chemistry & Electron High Technology Co., Ltd., China). A titanium layer of 500 nm was magnetron sputtered onto a 4 in. Si wafer. A working electrode with an active area of 8 mm × 16 mm was cut from this piece. The Ti layer was treated before use by the method described in Ref. [19] to enhance the adhesion between Ti and PPy film. A platinum wire was applied as a counter electrode. All potentials were referred to as saturated calomel electrode (SCE). To ensure the whole surface of the electrode covered by PPy nanowire network, a thin layer of PPy about 1 μm with conventional morphology was synthesized firstly on the Ti electrode in an electrolyte containing 0.14 M pyrrole and 0.07 M lithium perchlorate at 1.0 V for 150 s. Then PPy nanowire networks were grown for 600 s from 0.07 M lithium perchlorate, 0.20 M ATP and 0.15 M pyrrole on the surface of the first PPy layer at 1.0 mA cm⁻². Prior to the electropolymerization, the electrolyte solution was degassed with a nitrogen flow for 15 min. After polymerization, the working electrode was removed from the electrolyte and rinsed thoroughly with de-ionized water, and then dried in air at room temperature. For comparison, two kinds of cauliflower-like PPy were synthesized. The first kind of cauliflower-like PPy was prepared by galvanostatic method under the same conditions as above except for the absence of ATP. The second kind of cauliflower-like PPy was synthesized from 0.07 M lithium perchlorate, 0.20 M ATP and 0.15 M pyrrole at the current density of 1.6 mA cm⁻² for 375 s.

2.3. Characterization and measurement

The surface morphologies of the prepared PPy were examined by a scanning electron microscope (SEM, LEO1530, Germany) operated at 20 kV. Cyclic voltammetric (CV) measurements were made in 0.1 M NaCl solution between potentials of -0.6 V and 0.8 V (scan rate: 20 mV/s), with a LK2005A Electrochemical Workstation at 25 °C. The electrochemical impedance spectrum (EIS) of the PPy products was measured in 0.1 M NaCl solution at 25 °C with a EG&G263A potentiostat/galvanostat (Princeton Applied Research, USA) and a Model 5210 Lock-in Amplifier (Princeton Applied Research, USA) using 10 mV (rms) AC sinusoid signal at a frequency range from 100 kHz to 1 Hz. All the data reported are the average values of at least 3 samples from each experimental condition.

2.4. ATP release

2.4.1. External powered ATP release

The external powered ATP release from the PPy nanowire network was carried out in 5 mL NaCl solution (0.9%) stirred with a magnetic stirring bar at room temperature. A negative potential of -0.80 V was applied. Samples (2.5 mL) were taken at specific times from the release medium, analyzed for ATP content by UV/vis spectrophotometer (UV-2000, Unico (Shanghai) Instrument Co., Ltd.) at 257 nm, and replaced with an equal volume of fresh 0.9% NaCl solution.

2.4.2. Self-powered ATP release

For self-powered release, a thin layer of magnesium with thickness of 500 nm was magnetron sputtered with or without a mask on the surface of the nanowire network using the JS-3X-100B magnetron sputtering machine (Chuangweina Technology Co., Ltd., China). In order to obtain a patterned surface, the surface of the nanowire network was selectively exposed to magnetron sputtering atmosphere by using a mask containing square holes of 2.5 mm × 2.5 mm. Only the exposed regions were covered by

magnesium layer. As a result, PPy nanowire network with patterned magnesium layers on their surface was obtained after removal of the mask. Then the ATP release was conducted by immersing the magnesium-coated PPy nanowire network (patterned or non-patterned) vertically in the same release medium as above except that the external electrical stimulation was not applied.

3. Results and discussion

3.1. Synthesis of the PPy nanowire network

In our experiment, if PPy nanowire network was deposited directly on the Ti electrode, it could not form a continuous film. To circumvent this problem, a non-nanostructured thin PPy layer was synthesized first on the Ti electrode and it was found that this pre-layer significantly improves the formation of the continuous nanostructured film and the whole electrode surface was covered completely by the PPy nanowire networks. Although the exact mechanism is not clear, the presence of the pre-layer may improve the polymer growth by providing a more rough surface and thus higher surface area of the electrode.

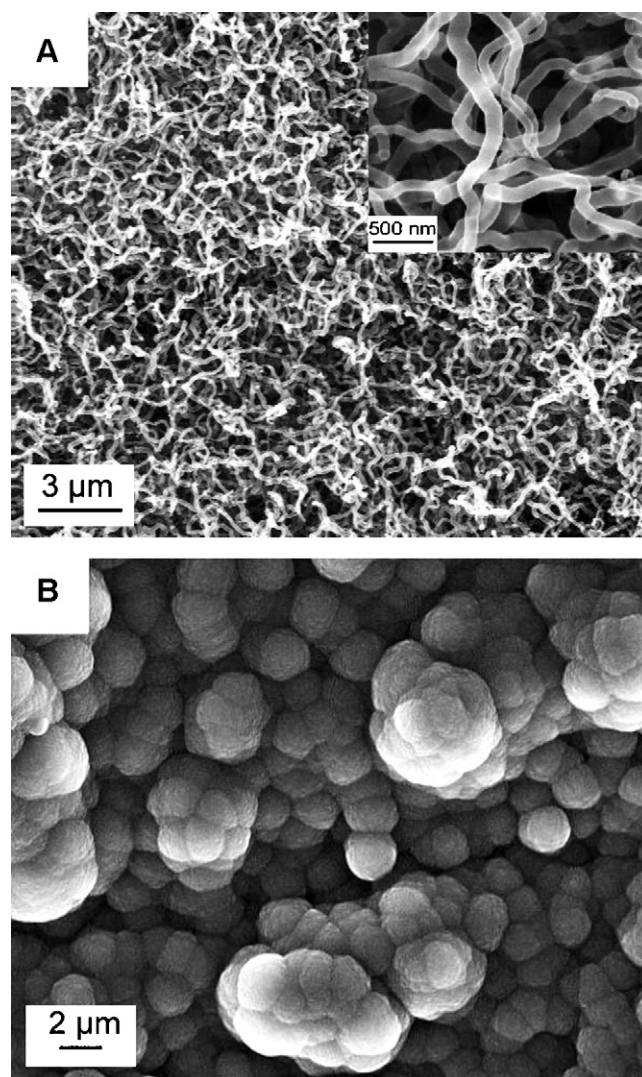


Fig. 1. SEM images of: (A) PPy nanowire networks (inset: high magnification image); (B) cauliflower-like PPy synthesized in the absence of ATP.

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