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Preparation of micropatterned polyaniline thin films with enhanced electrochromic properties by electrostatic field-assisted potentiostatic deposition

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ABSTRACT

Micropatterned polyaniline (mPani) thin films on indium tin oxide (ITO) conducting substrates are prepared by the electrostatic field-assisted potentiostatic deposition (ESaPD) and their electrochromic (EC) properties are investigated in this study. The mPani thin films can be directly electrodeposited onto the ITO substrates under the influence of a patterned electrostatic field induced by a micropatterned electrostatic film through contact electrification. It is found that the mPani thin film has faster coloring/bleaching responses, small enhancement in transmittance modulation ability and higher coloration efficiency than the non-patterned Pani thin film prepared with the same deposition time. Electrochemical impedance spectroscopy analysis indicates that the charge-transfer resistance of the mPani thin film is smaller than that of the non-patterned Pani thin film, and which could be the reason for the enhancement of the EC properties of the mPani thin film.

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1. Introduction

Electrochromic (EC) materials have the ability to change their absorbance properties of electromagnetic radiations upon the application of external potentials [1]. The theory of electrochromism was suggested by Platt [2] in 1961 and was first demonstrated by Deb [3] in 1969. Polyaniline (Pani) is a well-known polymeric EC material with multiple color states and stable chemical and electrochemical properties, and its EC properties as well as applications have been extensively studied [1,4]. Pani can reversibly change its color between light yellow of the fully reduced state (Leucoemeraldine base, LB), green of the partially oxidized state (Emeraldine base, EB), and blue of the fully oxidized state (Pernigraniline base, PB) through its electrochemical redox reactions. Pani thin films can be prepared by electrochemical polymerization [5] on conducting substrates. The modifications of Pani nanostructures [6–8] or the developments of Pani composites with other materials [9–11] for improving the EC performances have also being widely explored. However, relatively few studies were concerning about the EC performances of micropatterned Pani (mPani) thin films.

Conventional methods for the preparation micropatterned thin films include photolithography [12], soft lithography [13], templating [14] or aqueous chemical growth [15]. Strategies of alternating the surface properties of the substrate such as hydrophobicity [16], photoconductivity [17], or static electricity [18] for deriving micropatterned thin films have also been studied. Most of these methods involved multiple pretreatment steps of the substrate. A novel electrostatic field-assisted potentiostatic deposition (ESaPD) method with one simple substrate pretreatment process has been demonstrated for the preparation of micropatterned inorganic thin film on indium tin oxide (ITO) conducting glass in our previous study [19]. By attaching a micropatterned electrostatic film (ESF) onto an ITO substrate through the contact electrification phenomenon [20], a patterned electrostatic field could be induced upon the removal of the ESF, and a micropatterned thin film could be directly electrodeposited on the ITO surface under the influence of the electrostatic field.

In this study, mPani thin films were prepared on ITO substrates using the ESaPD method. EC properties including the coloring/bleaching response time, transmittance modulation ($\Delta T\%$), and coloration efficiency (CE) of these mPani thin films were investigated and compared with the non-micropatterned Pani thin films prepared with the same deposition parameters. Electrochemical impedance spectroscopy (EIS) analysis was employed to characterize the kinetic parameters of the electrochemical reactions of these thin films.

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2. Materials and methods

Aniline (99.5%) and hydrochloric acid (HCl, 37%) were purchased from Acros. Potassium chloride (KCl, 99.9%) was obtained from Shimakyu's Pure Chemicals. All chemicals were used as received without further purification. Deionized water was used throughout the electrochemical experiments. ITO conducting glass (7 Ω/sq) was purchased from Solaronix. ITO conducting glass was sequentially sonicated in diluted detergent solution, acetone and deionized water each for 5 min, dried by nitrogen flow and then kept in a desiccant case before use. Electrostatic film (PE-1101E) was obtained from Youlen Technology Co,. Ltd. Optical microscope (OM) image of the ESF is shown in Fig. 1. The ESF consisted of a polyethylene (PE) sheet with disk-shaped polyacrylate (PA) bumps orderly arranged on its surface. The height and diameter of the bumps were 5 μm and 220 μm, respectively.

Surface morphology of the thin films was observed using a scanning electron microscope (SEM, Carl Zeiss, Leo 1530). Digital images of the thin films were obtained using an optical microscope (Olympus, BX51). A conventional three-electrode system with a potentiostat/galvanostat (CH Instrument, CHI-760D) was employed for the electrochemical experiments. A platinum coil was used as the counter electrode and the reference electrode was a Ag/AgCl electrode. The working electrode was an ITO glass for the Pani electrodeposition and was a mPani or Pani-coated ITO glass for other electrochemical experiments. The area of the working electrode was 1×2 cm^2. The electrolyte for cyclic voltammetry (CV), double-potential-step (DPS) and electrochemical impedance spectra (EIS) experiments was an aqueous solution containing 0.1 M KCl and 0.01 M HCl. An ultraviolet–visible spectrophotometer (Thermo, EVO300PC) was used for the in-situ measurements of the absorbance changes during the electrochemical reactions of the mPai and Pani thin films.

The procedure for the preparation of a mPani thin film is described as follows: The ESF was attached onto the ITO surface through contact electrification for at least 7 days and was peeled off right before the electrodeposition process. Pani was then potentiostatically deposited in an aqueous solution containing 2.0 M HCl and 1.0 M aniline. A potential of 0.8 V (vs. Ag/AgCl) was applied to the ESF-pretreated ITO substrate for various deposition times to deposit Pani. The mPani thin films prepared with the deposition time of 80 s, 100 s, 120 s, 140 s, and 160 s were named as mPani-80, mPani-100, mPani-120, mPani-140, and mPani-160 thin films, respectively. Non-micropatterned Pani thin films (Pani-80, Pani-100, Pani-120, Pani-140, and Pani-160 thin films) were also prepared using the same procedures on pristine ITO substrates for comparisons.

It may not be suitable for real fabrication if the preparation of a micropatterned film takes several days. The strength of an electrostatic field generated by contact electrification depends on the two materials brought in contact. According to the triboelectric series, it is expect that greater electrostatic field could be generated in a shorter time by contacting an ITO surface with other materials such as polyurethanes or polyvinylchloride. Studies in reducing the preparation time are ongoing in our laboratory.

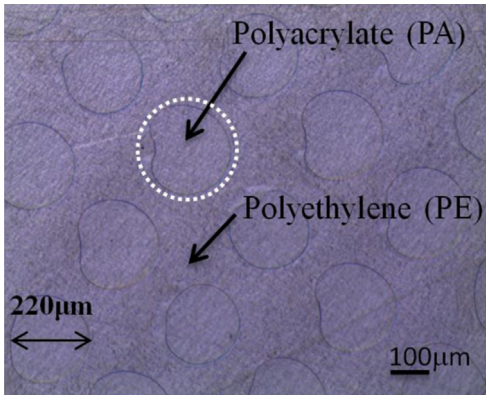

Fig. 1. OM images of the ESF film.

3. Results and discussion

3.1. Procedures and mechanisms of the ESaPD

The mPani thin films were prepared using the ESaPD method. Procedures of the ESaPD and the proposed mechanism are illustrated in Fig. 2. The ESF was firstly attached onto the ITO surface through contact electrification. The ESF was stored in roll and in that way the PA bumps were in contact with the backside of the PE sheet. According to the tribo-electric series reported in previous studies [21,22], equal amount of positive and negative charges could be developed on the PA and the PE sides, respectively. Therefore there were positive charges initially on the PA bumps surface. When the positive-charged PA bumps were brought in contact with the ITO surface, negative charges could be attracted and accumulated at the ITO side of the PA/ITO interface. These negative charges should induce an equal amount of positive charges gathered at the opposite side of the ITO layer (the ITO side of the ITO/glass interface). Glass is an electret material [23] which could retain quasi-permanent charge separations. The positive charges at the ITO side of the ITO/glass interface might induce the formation of charge separations inside the glass, as shown in Fig. 2. When the ESF was removed from the ITO surface, a positive electrostatic field was thus built up by the positive charges at ITO side of the ITO/glass interface. The surface areas that have been contacted with the PA bumps were thus experiencing the positive electrostatic field. There should be no local negative charges accumulated on the ITO surface since it is a conductor. The locally distributed electrostatic field on the ITO surface could affect the potentiostatic deposition process and a mPani thin film might be obtained. The ITO surface areas with and without the influence of the underlying positive electrostatic field were designated as A_+ and A_0, respectively (Figs. 2 and 3(f)). A_+ was the area that has been attached with the PA bumps, and A_0 was the pristine ITO surface area. The electrodeposition of Pani is an oxidative process

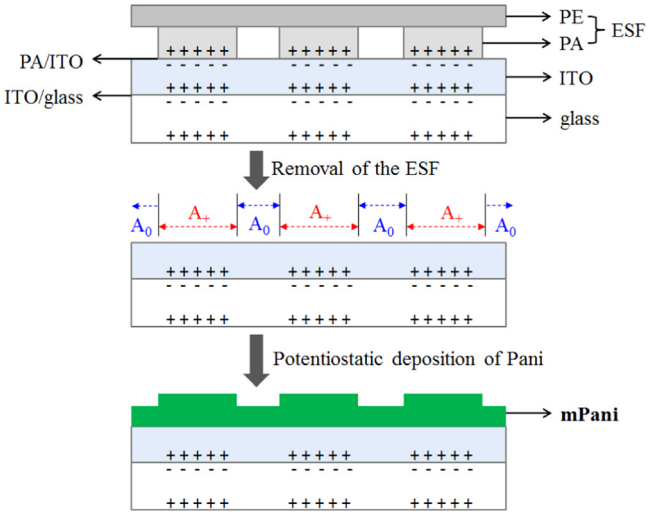

Fig. 2. Illustrations of the procedures and the proposed mechanism of the ESaPD of a mPani thin film on an ITO substrate.

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