

## FOCUS REVIEW

# Fabrication of gradient polymer surfaces using bipolar electrochemistry

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This report focuses on recent developments using bipolar electrochemistry as an effective tool for the fabrication of gradient polymer surfaces. The electrochemical doping and reactions of conducting polymers under an applied potential distribution using bipolar electrodes have been carried out to prepare conducting polymers with composition gradients. Indirect electrolysis using an electrogenerated metal catalyst on bipolar electrodes successfully afforded gradually modified polymer surfaces and gradient polymer brushes using vinyl monomers. The newly designed cylinder bipolar electrode system is available for site-selective applications of electric potentials, which produced electrochemical patterning of conducting polymer films.

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## INTRODUCTION

An in-plane gradient substrate surface can impart a variety of chemical and physical properties. Gradient surfaces have an important role in biological system, as well as for the screening and sensing of materials. To prepare gradients on a substrate, many approaches have been reported for gradually modifying the surface such as using chemical or physical bonding, treating precoated materials with gradient stimuli and so on, as previously reported in related reviews.<sup>1,2</sup> Electrochemical methods utilizing anisotropic fields driven by a pair of electrodes result in versatile functional gradients that are easily controlled by driving potentials.<sup>3</sup>

Among the electrochemical techniques for preparing gradient surfaces, bipolar electrochemistry is a powerful tool for generating in-plane potential gradients at the electrode surface.<sup>4,5</sup> In an electrolytic cell containing driving electrodes and an isolated conducting substrate, a bipolar electrode (BPE) is formed when the external electric field from the driving electrodes is sufficiently strong to produce both anodic and cathodic reactions in a low concentration of electrolyte. When the anodic and cathodic surfaces coexist on the same BPE plane, a potential distribution appears on the BPE. To exploit this phenomenon for material applications, many interesting approaches involving reactions of small substrates being fixed<sup>6,7</sup> or deposited<sup>8–10</sup> on the BPE have been reported. A typical potential distribution is linear and reflects the linearly applied electric field. However, we have successfully controlled the potential slope on the BPE by designing a cell configuration that includes insulating walls (Figure 1).<sup>11,12</sup>

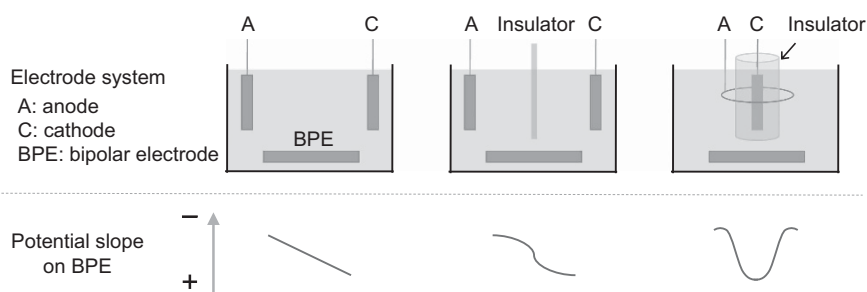
Herein, the use of this type of potential distribution is demonstrated for the fabrication of gradients on polymeric materials. Thus, a redox active, conducting polymer film was employed as a substrate on a BPE to directly or indirectly reflect the potential gradients. In addition, surface-initiated polymerization using an electrochemically generated metal catalyst in a gradient manner was also investigated.

## RESULTS AND DISCUSSION

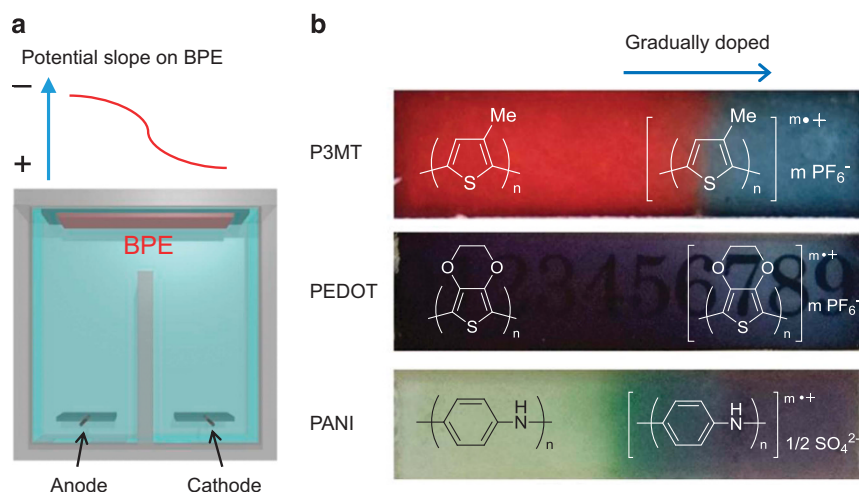
### Gradient doping of conducting polymers

Conducting polymers can produce electron transfer on an electrode surface to form polarons and bipolarons in the repeating structure (commonly known as doping), producing considerable variations in the physical properties of the polymer itself and imparting features, such as drastic color changes and electrical conductivity.<sup>13,14</sup> The charges generated along the polymer must be compensated by the addition of neighboring ions derived from the electrolyte (dopants). The doping process is typically reversible (that is, the application of the opposite potential to the doped polymer returns it to its neutral state).

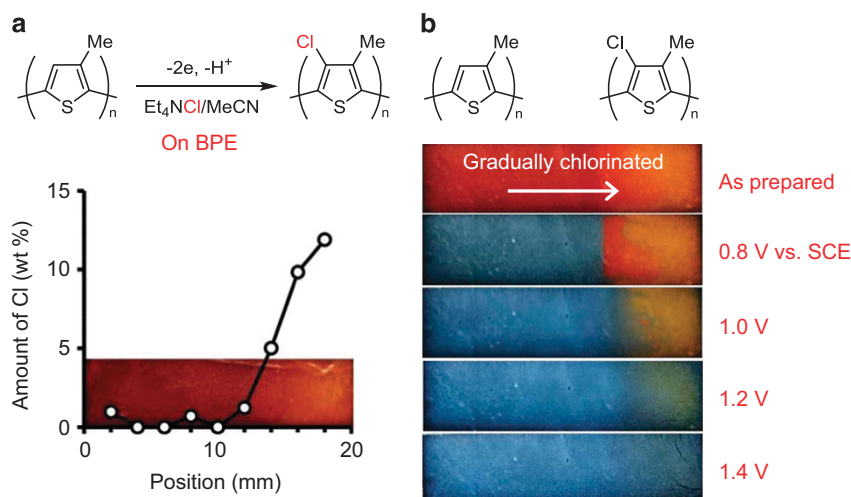
The application of gradient potentials generated on a BPE to conducting polymers was employed to obtain composition-gradient materials with different doping levels across the film. First, a poly(3-methylthiophene) (P3MT) film was prepared on an indium tin oxide (ITO) working electrode using potential sweep electropolymerization, and then, this film was placed into a U-type cell equipped with a Pt anode and a Pt cathode connected to a constant current power supply containing 5 mM Bu<sub>4</sub>NPF<sub>6</sub>/MeCN (Figure 2a).<sup>15</sup> On the BPE, the side facing the cathode acted as an anodic surface, and the other side acted as a cathodic surface with a sigmoidal profile. When the potential difference between both poles was above the threshold for the anodic reaction (electrochemical doping) and cathodic reaction (reduction of contaminating water and/or oxygen), these reactions simultaneously occurred on the BPE. The P3MT, which was initially a red-colored film, turned blue at the anodic pole of the BPE during the passage of constant current (Figure 2b). The current that passed between the driving electrodes changed the steepness of the potential slope on the BPE, indicating the significant change in the doped area of the P3MT film.<sup>11</sup> Similarly, the poly(3,4-ethylenedioxythiophene) (PEDOT) film on the BPE exhibited a drastic change in color from deep blue to being rather transparent on its anodic side. For polyaniline



**Figure 1** Diagrams of a typical BPE apparatus without or with an insulating wall and the potential slope generated on the BPE in each case. A full color version of this figure is available at *Polymer Journal* online.



**Figure 2** (a) U-type electrolytic cell containing a driving anode and cathode pair and BPE covered with a conducting polymer film. The potential slope on the BPE is also illustrated. (b) Photographs of the P3MT, PEDOT and PANI films gradually doped on the BPE.

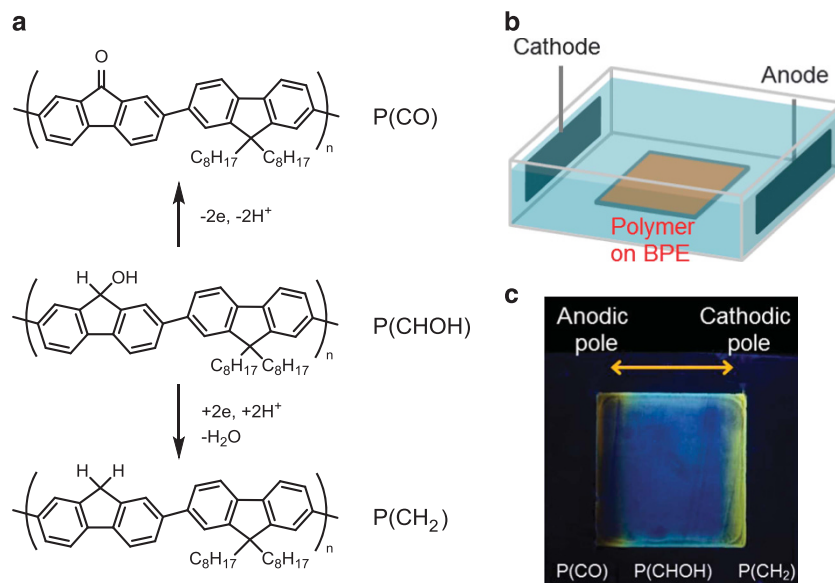


**Figure 3** (a) Photograph of the gradually chlorinated P3MT film on the BPE. The profile showing the amount of chlorine atoms across the film is embedded. (b) Photographs of the gradually chlorinated P3MT film in the course of anodic doping at different potentials.

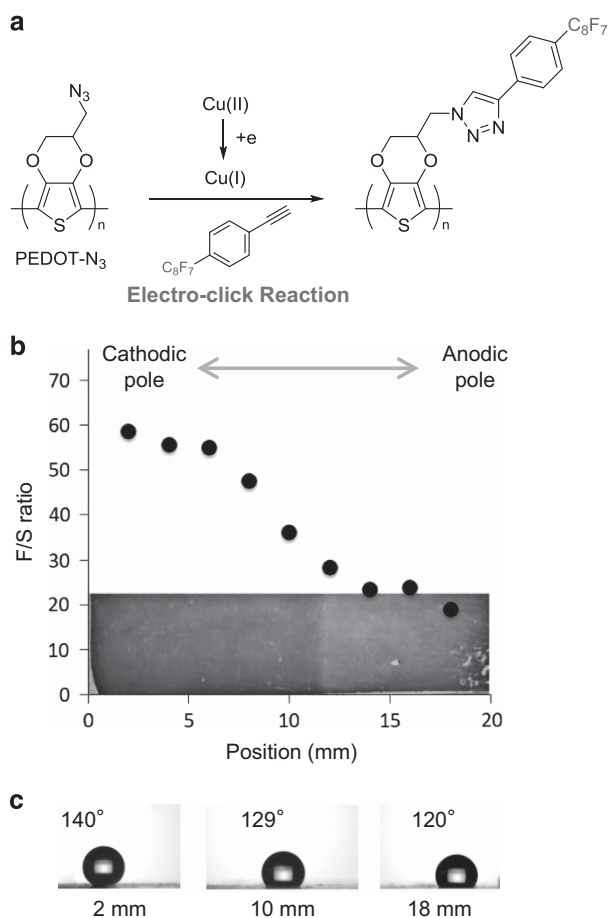
(PANI) in 5 mM  $H_2SO_4$ , a multicolored gradient was observed on the film, depending on the applied potential on the BPE. The absorption spectrum at each position was compared with that of the authentic samples doped under potentiostatic conditions. Therefore, the potential distribution generated on the BPE was successfully estimated.

### Gradient reactions of conducting polymers

When a doped state of a conducting polymer is unstable, over-oxidation or overreduction (that is, undesirable reactions with nucleophiles or electrophiles, respectively) may occur, resulting in the formation of a non-conducting polymer.<sup>16</sup> Therefore, the



**Figure 4** (a) Possible anodic and cathodic reactions of P(CHOH). (b) Schematic illustration of the setup used for bipolar electrolysis of the P(CHOH) film. (c) Photograph of the obtained conducting polymer film under UV irradiation ( $\lambda = 365$  nm), exhibiting a gradient composition.



**Figure 5** (a) Scheme for the electro-click reaction of PEDOT-N<sub>3</sub> and a perfluorinated alkyne derivative using an electrogenerated Cu(I) species. (b) The weight ratio (F/S) of the modified film at each position of the BPE determined by EDX. (c) Contact angle of water droplets (1  $\mu$ l) at the different positions of the modified film. A full color version of this figure is available at *Polymer Journal* online.

appropriate choice of 'reactive dopants' can lead to selective reactions that afford a functionalized polymer. Based on this concept, we have developed electrochemical polymer reactions that are initiated by electrochemical doping followed by chemical reactions.<sup>17–20</sup>

The electrochemical polymer reactions on a BPE are possible because of the use of reactive dopants in the gradually doped state of conducting polymers. Therefore, the oxidative chlorination of P3MT on an ITO plate, which acts as a BPE, was investigated in 5 mM Et<sub>4</sub>NCl/MeCN (Figure 3a).<sup>21</sup> After the passage of constant current between the driving electrodes, the ITO covered with the product polymer was dedoped at  $-0.3$  V (vs SCE) to completely remove the dopant. The EDX results provided clear evidence of the successful introduction of chlorine atoms into the film on the anodic part, which reflects the potential gradient of the BPE. The chlorine substitution on the poly(thiophene) derivatives often imparts a tolerance to oxidation because it lowers the highest occupied molecular orbital (HOMO) energy level.<sup>22,23</sup> Therefore, the chlorine-gradient in the P3MT film corresponds to an intrinsic energy gradient across the film. Anodic doping of the prepared chlorine-gradient film was also performed in 0.1 M Bu<sub>4</sub>NPF<sub>6</sub>/MeCN (Figure 3b). When the gradient film was charged at 0.8 V (vs SCE), only the non-chlorinated area was doped and subsequently exhibited blue color. The interface of the doped and undoped polymer was gradually shifted to the chlorine-rich area as a function of the applied potential. The entire polymer film was doped when charged at 1.4 V (vs SCE).

#### Parallel polymer reactions on a BPE

The parallel electrochemical polymer reactions can be performed with anodic oxidation and cathodic reduction on both electrodes simultaneously when an oxidizable and reducible conducting polymer is used. A polyfluorene derivative bearing a 9-hydroxyfluorene moiety as an electroactive unit (P(CHOH)) provides a polyfluorenone derivative (P(CO)) by oxidation and a polyfluorene derivative (P(CH<sub>2</sub>)) by reduction (Figure 4a).<sup>24</sup> The combination of bipolar electrochemistry and parallel polymer reactions is very interesting because the divergent transformation of the 9-hydroxyfluorene-based precursor polymer can be achieved on both poles of a BPE. Therefore, a multi-composition-

gradient film is expected as the product. A boron-doped diamond plate covered with P(CHOH) was placed between driving stainless electrodes in the setup shown in Figure 4b, and a constant current was passed in 5 mM Et<sub>4</sub>NOTs/2-propanol. After electrolysis, under UV irradiation, the emission color of the polymer film consisted of a gradation of color from yellow to blue to dark orange from the cathodic pole to the anodic pole (Figure 4c). The gradient polymer film was peeled off the boron-doped diamond plate with adhesive tape and divided into three pieces. The PL spectrum of each section corresponded well to the spectra of the polymers. This parallel bipolar electrolysis successfully provided a new composition-gradient film.

### Electro-click modification of conducting polymer surface

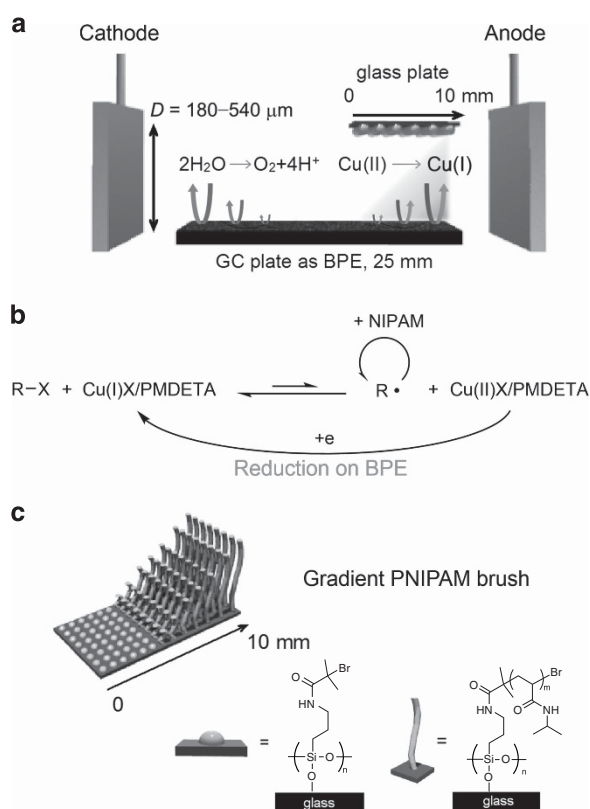
The electro-click reaction, which utilizes cathodically generated Cu(I) species for azide-alkyne cycloaddition (click reaction), is a powerful tool for surface modification of an electrode.<sup>25,26</sup> Electropolymerization of 3,4-(1-azidomethylethylendioxy)thiophene (EDOT-N<sub>3</sub>) was carried out to prepare its polymeric film (PEDOT-N<sub>3</sub>) on an ITO electrode. After dedoping, the neutral polymer was obtained as a blue film. The polymer film, which was fixed on the ITO substrate, was placed into a U-type electrolytic cell containing a mixed solution of water/BuOH (v/v = 2:1) containing copper sulfate (10 mM) and an alkyne with a perfluoroalkyl group. The copper sulfate acted as the supporting electrolyte and the source of Cu(I) ion to catalyze the azide-alkyne cycloaddition (Figure 5a). After bipolar electrolysis, the gradual introduction of the perfluoroalkyl group was observed by EDX. The fluorine-content profile revealed that the electrogenerated

Cu(I)-mediated click reaction reflected the potential profile on the BPE (Figure 5b).<sup>27</sup> Then, the surface property of the gradually perfluoroalkylated PEDOT was investigated by measuring the static contact angle of a water droplet on the film. As expected, the contact angle at the fluorinated surface was significantly larger than that at the pristine surface (Figure 5c). The gradual decrease in the contact angle from the cathodic surface to the anodic surface corresponded well to the modification level using the electro-click reaction on the BPE.

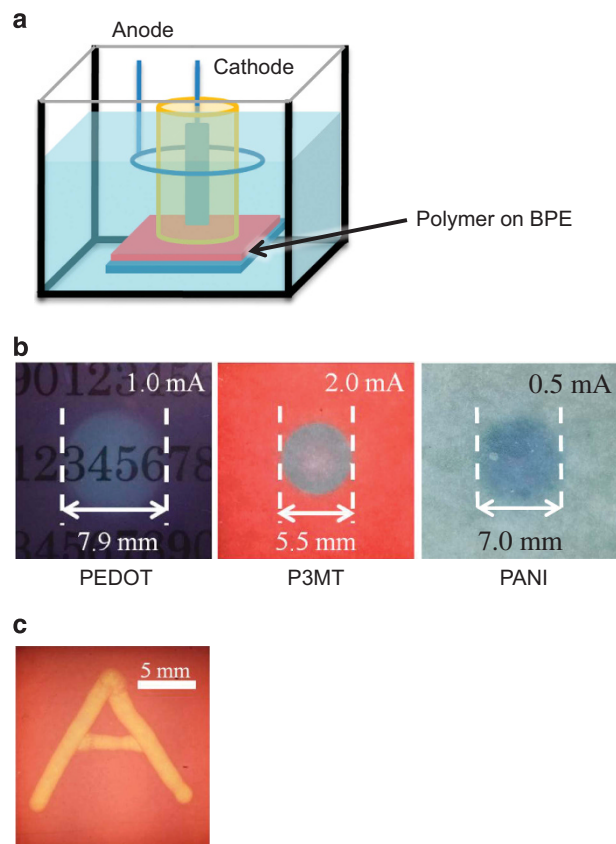
### Fabrication of gradient polymer brushes

The electrochemically generated Cu(I) species can also be used as a catalyst for atom transfer radical polymerization (ATRP) of vinyl monomers. This electrochemically mediated ATRP (eATRP) has attracted much attention because controlled polymerization can be achieved by changing the electrochemical parameters.<sup>28,29</sup> The bipolar electrochemistry was employed to generate a concentration gradient with Cu(I), which was derived from the reduction of Cu(II) at the cathodic pole of a BPE for the surface-initiated ATRP of monomers. This process resulted in the formation of polymer brushes with an in-plane height gradient.<sup>30</sup>

We prepared an eATRP experimental apparatus by sandwiching a glassy carbon plate that acts as the BPE with an initiator-modified glass plate with the incorporation of a microgap (*D*) (Figure 6a). When a sufficient voltage was applied between the driving electrodes, an electric field was generated in the cell, generating a BPE at which simultaneous anodic oxidation of water and cathodic reduction of



**Figure 6** (a) Illustration of the setup for surface-initiated eATRP using bipolar electrochemistry. (b) General scheme for eATRP of the NIPAM monomer using a Cu catalyst. (c) Representation of a gradient poly(NIPAM) brush propagated from the initiator-modified glass substrate. A full color version of this figure is available at *Polymer Journal* online.



**Figure 7** (a) Schematic illustration of the setup for bipolar patterning. (b) Photographs of the conducting polymer films showing the site-selectively doped PEDOT, P3MT and PANI using a cylinder with a 6.0 mm diameter. (c) Photograph of the line-patterned P3MT film by anodic chlorination using a cylinder with diameter of 1.0 mm.

copper(II) species occurred. These reactions were influenced by the potential distribution on the BPE. Therefore, the concentration gradient of the Cu(I) catalyst generated at the BPE produced a reaction field over which the  $[\text{Cu(I)}]/[\text{Cu(II)}]$  concentration ratio varied slightly with position. In this manner, the polymerization rate gradually changed with position according to the eATRP reaction mechanism (Figure 6b). As a result, polymer brushes with three-dimensional gradients were successfully produced (Figure 6c).

For the polymerization of *N*-isopropylacrylamide (NIPAM) on a glass substrate by eATRP via bipolar electrochemistry, a 3.2 M NIPAM solution in  $\text{H}_2\text{O}/\text{MeOH}$  (1/1) with  $\text{CuCl}_2$  (2 mM) was used, and *N,N,N',N',N''*-pentamethyl-diethylenetriamine was used as the electrolyte. The microgap, *D*, was 360  $\mu\text{m}$  in these trials. After applying a  $\Delta V_{\text{BPE}}$  value of 1.4 V for 60 min under ambient conditions, the glass plate was washed with solvent to remove physically adsorbed compounds. Film thickness measurements at various positions were performed with a stylus-type tester to generate a height profile. A gradient increase was observed when moving toward the cathodic edge of the BPE, which was due to the gradient change in the degree of polymerization. The height of the polymer brush increased in a linear manner as the polymerization time increased, indicating that the surface-initiated eATRP proceeded in a living-like manner. The polymer brush film thickness can be tuned by changing various parameters, such as the applied voltage ( $\Delta V_{\text{BPE}}$ ) and the microgap (*D*). When a higher voltage was applied to the BPE, the film became slightly thicker in an increasingly steep manner. A smaller gap afforded a thicker brush film along with expanding the modification area.

#### Site-controlled application of potential on a BPE

As previously described, site-controlled potential application on a BPE is possible by designing the setup for electrolysis.<sup>12</sup> The spot-bipolar configuration is shown in Figure 7a. The conducting polymer film was prepared on an ITO electrode by electropolymerization and used as the BPE in a container equipped with an external cathode wire and an anode ring separated by a plastic shielding cylinder. When current flowed via the BPE in a solution containing a low electrolyte concentration, a new anodic area appeared under the driving cathode wire along with a surrounding cathodic area with an interface gradient. The employed cylinders had internal and external diameters of several millimeters. The distance between the cylinder and the BPE was maintained at 1 mm. Figure 7b shows photographs of the PEDOT, P3MT and PANI films that were doped with the bipolar device under a constant current.<sup>12,31</sup> A clear spot appeared in the anodic pattern of the BPE. This local color change completely recovered when the entire substrate was treated with cathodic electricity (dedoping). When a nucleophilic supporting salt (5 mM  $\text{Et}_4\text{NCl}/\text{MeCN}$ ) was used, a yellow circle on the P3MT film was irreversibly formed due to electrochemical chlorination at the 4-position of the repeating thiophene unit. Similarly, the line patterning based on the chlorination of P3MT was successfully demonstrated by moving the cylinder system (Figure 7c).

Using a similar setup, the Cu(I) species was successfully and site-selectively generated on a mesh BPE and used for the eATRP of the NIPAM monomer on an initiator-modified glass plate. Therefore, the poly(NIPAM) brush formed on the glass plate had a circular shape that corresponded to the size of the cylinder used. The patterned area possessed a water droplet at room temperature. However, at 60 °C, the surface became hydrophobic due to the phase separation of poly(NIPAM) in water.<sup>30</sup>

#### SUMMARY

The fabrication of gradient polymer surfaces using bipolar electrochemistry was investigated. The gradient electrochemical doping and subsequent chemical reactions of conducting polymers on a BPE were successfully demonstrated. The concentration gradient of an electrogenerated metal catalyst was also effective for the post-functionalization of a conducting polymer surface and surface-initiated eATRP of vinyl monomers. In addition, the site-selective application of electric potentials on a BPE is a useful approach for electrochemical patterning applications. The combination of electrochemical reactions of polymeric materials and bipolar electrochemistry was very effective for creating gradient functionalities designed by the potential distribution on the BPEs.

#### CONFLICT OF INTEREST

The authors declare no conflict of interest.

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