

## Rational Design of Electrochemiluminescent Devices

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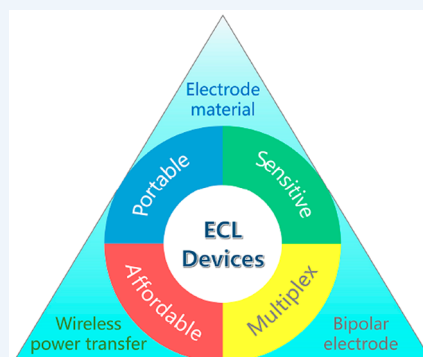
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**CONSPECTUS:** Electrochemiluminescence (ECL) is a light-emitting process which combines the intriguing merits of both electrochemical and chemiluminescent methods. It is an extensively used method especially in clinical analysis and biological research due to its high sensitivity, wide dynamic range, and good reliability. ECL devices are critical for the development and applications of ECL. Much effort has been expended to improve the sensitivity, portability, affordability, and throughput of new ECL devices, which allow ECL to adapt broad usage scenarios.

In this Account, we summarize our efforts on the recent development of ECL devices including new electrodes, ECL devices based on a wireless power transfer (WPT) technique, and novel bipolar electrochemistry. As the essential components in the ECL devices, electrodes play an important role in ECL detection. We have significantly improved the sensitivity of luminol ECL detection of  $H_2O_2$  by using a stainless steel electrode. By using semiconductor materials (e.g., silicon and  $BiVO_4$ ), we have exploited photoinduced ECL to generate intense emission at much lower potentials upon illumination. For convenience, portability, and disposability, ECL devices based on cheap WPT devices have been designed. A small diode has been employed to rectify alternating current into direct current to dramatically enhance ECL intensity, enabling sensitive ECL detection using a smart phone as a detector. Finally, we have developed several ECL devices based on bipolar electrochemistry in view of the convenience of multiplex ECL sensing using a bipolar electrode (BPE). On the basis of the wireless feature of BPE, we have employed movable BPEs (e.g., BPE swimmers and magnetic rotating BPE) for deep exploration of the motional and ECL properties of dynamic BPE systems. To make full use of the ECL solution, we have dispersed numerous micro-/nano-BPEs in solution to produce intense 3D ECL in the entire solution, instead of 2D ECL in conventional ECL devices. In addition, the interference of ECL noise from driving electrodes was minimized by introducing the stainless steel with a passivation layer as the driving electrode. To eliminate the need for the fabrication of electrode arrays and the interference from the driving electrode and to decrease the applied voltage, we develop a new-type BPE device consisting of a single-electrode electrochemical system (SEES) based on a resistance-induced potential difference. The SEES is fabricated easily by attaching a multiperforated plate to a single film electrode. It enables the simultaneous detection of many samples and analytes using only a single film electrode (e.g., screen-printed electrode) instead of electrode arrays. It is of great potential in clinical analysis especially for multiple-biomarker detection, drug screening, and biological studies. Looking forward, we believe that more ECL devices and related ECL materials and detection methods will be developed for a wide range of applications, such as *in vitro* diagnosis, point-of-care testing, high-throughput analysis, drug screening, biological study, and mechanism investigation.



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- Qi, L.; Xia, Y.; Qi, W.; Gao, W.; Wu, F.; Xu, G. Increasing Electrochemiluminescence Intensity of a Wireless Electrode Array Chip by Thousands of Times Using a Diode for Sensitive Visual Detection by a Digital Camera. *Anal. Chem.* **2016**, 88, 1123–1127.<sup>2</sup> A wireless power supply array chip was developed for ECL detection with a diode that could rectify the alternating current and increase the ECL intensity by thousands of times.
- Gao, W.; Muzyka, K.; Ma, X.; Lou, B.; Xu, G. A single-electrode electrochemical system for multiplex electrochemiluminescence analysis based on a resistance induced potential difference. *Chem. Sci.* **2018**, 9, 3911–3916.<sup>3</sup> A special type of bipolar electrode system for multiplex ECL analysis was designed on the basis of the resistance-induced potential difference of ITO glass.

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- Zhao, Y.; Yu, J.; Xu, G.; Sojic, N.; Loget, G. Photoinduced Electrochemiluminescence at Silicon Electrodes in Water. *J. Am. Chem. Soc.* **2019**, *141*, 13013–13016.<sup>4</sup> A photo-electrochemiluminescence system was developed, allowing the production of light from photogenerated holes at silicon electrodes at remarkably low potentials in water.

## 1. INTRODUCTION

Electrochemiluminescence (ECL), also called electrogenerated chemiluminescence, is a luminescence process whereby excited states of luminophores are electrochemically generated in the vicinity of the electrode surface and then relax to a lower state, leading to light emission.<sup>5–7</sup> By dexterously combining the electrochemical and chemiluminescence strategies, ECL inherits the fascinating merits of both techniques, such as high sensitivity, a wide dynamic range, and easy multiplex imaging analysis of chemiluminescence as well as good stability and tunability of electrochemical methods. The appealing temporal and spatial control of light emission makes the ECL technique a very powerful platform for both sensing and imaging.<sup>8–10</sup> In fact, ECL has been extensively studied in many areas of analytical chemistry including immunoassays,<sup>10,11</sup> environmental detection,<sup>12</sup> aptasensors,<sup>13</sup> and so on, benefiting from the exploration of novel luminophores,<sup>14,15</sup> coreactants,<sup>16,17</sup> accelerators,<sup>18</sup> electrode materials,<sup>19,20</sup> and innovative devices<sup>21,22</sup> in the ECL detection process.

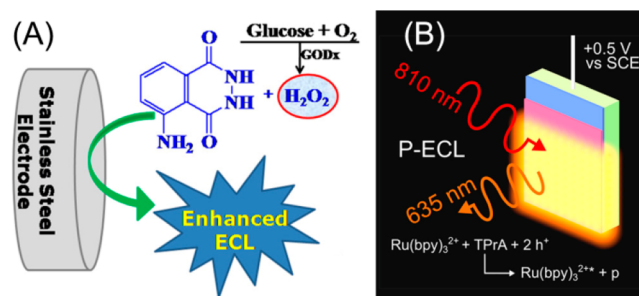
In recent years, with the rise of materials science and the generalization of electric technology, the design of ECL devices and the integration of new functions are more diversified than ever before. For example, scientists have invented diverse ECL devices for point-of-care testing,<sup>22</sup> cell microscopic imaging,<sup>23</sup> electrochemical imaging,<sup>24</sup> and wearable sensors<sup>25</sup> based on the superior properties of ECL. In this Account, we introduce our efforts on the exploration of novel ECL devices in terms of new electrodes,<sup>4,26,27</sup> the wireless power transfer technique,<sup>1,2,28</sup> and novel bipolar electrochemistry systems.<sup>3,28–34</sup> Also, the detailed background information and some important work from other groups in these areas are reviewed. In spite of the wide report on ECL devices, there are tough challenges that demand comprehensive design schemes and further endeavors. This Account aims to stimulate and inspire the embracing of ECL methods, original strategies, and booming new technologies and contribute to the endeavors for the development of ECL devices in analytical chemistry.

## 2. EXPLORATION OF NOVEL ELECTRODES FOR ECL DEVICES

The electrodes are essential components of ECL devices. Indeed, ECL is triggered by an initial electrochemical reaction at the electrode surface and subsequent chemical reactions in the vicinity of the electrode. Thus, the performance of ECL methods depends directly on the materials of electrodes.<sup>20</sup> In this section, we introduce some electrodes with unique performance.

Luminol is one of most popular ECL luminophores because of its superior features such as low cost, high ECL quantum efficiency in aqueous solutions, and broad spectrum of applications. It has been used in the direct detection of H<sub>2</sub>O<sub>2</sub>, the indirect detection of enzymes and their substrates through the detection of H<sub>2</sub>O<sub>2</sub>, and so on.<sup>7,8,10,11</sup> Therefore, it is important to find electrodes that can realize the sensitive determination of H<sub>2</sub>O<sub>2</sub>. However, almost all bare electrodes have limited sensitivity, and chemically modified electrodes have

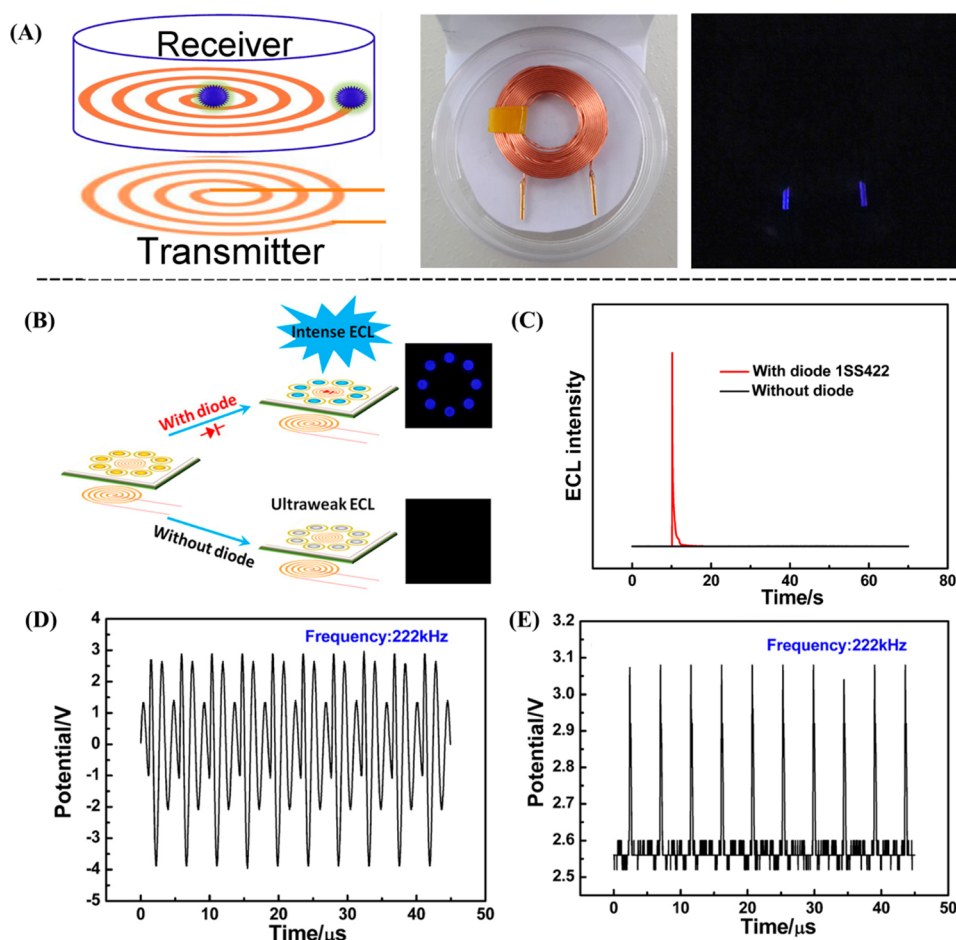
the disadvantage of relatively complex modification. Recently, we have first exploited luminol/H<sub>2</sub>O<sub>2</sub> ECL on a stainless steel electrode (Figure 1A).<sup>26</sup> The stainless steel electrode presents a



**Figure 1.** (A) Schematic diagram of the stainless steel electrode for ECL detection using the luminol/H<sub>2</sub>O<sub>2</sub> system. (B) Scheme of photoinduced ECL at silicon electrodes protected by a few-nanometers-thick SiO<sub>x</sub> and Ni-stabilizing thin films. Reproduced with permission from refs 26 and 4. Copyrights 2017 and 2019, respectively, American Chemical Society.

thin passivation layer on its surface. It can be regarded as a unique passivation-layer-modified electrode. This thin passivation layer endows the stainless steel electrode with some advantages over other metal electrodes (e.g., Pt, Au), such as a wide electrochemical window. We surprisingly found that a type-304 stainless steel electrode can significantly improve the sensitivity of the ECL detection of H<sub>2</sub>O<sub>2</sub> with a wide linear range of H<sub>2</sub>O<sub>2</sub> from 1 to 1000 nM. Compared with other bare electrodes, the stainless electrode improved the sensitivity by about 3 orders of magnitude with the help of the thin passivation layer formed on the stainless steel electrode surface. Glucose oxidase (GOx) can catalyze the reaction of glucose oxidation to generate H<sub>2</sub>O<sub>2</sub> in the presence of oxygen. On the basis of this enzymatic reaction, the stainless steel electrode was then successfully applied for the determination of glucose with a wider linear range and a LOD of 76 nM which is more sensitive than other luminol-based ECL glucose biosensors. Another attractive feature of this stainless steel electrode is that it can spontaneously form a thin passivation layer by instant oxidation in air after polishing and avoid the complex and time-consuming modification process in other chemically modified electrodes. In addition, its low cost, flexibility, and durability make the stainless steel electrode promising in disposable sensors and wearable sensors.

Semiconductor materials are mostly applied in the photo-electrochemistry (PEC) due to the high photoelectric efficiency, yet they have been seldom used in ECL. We have recently developed several photoinduced ECL (P-ECL) systems based on semiconductor electrodes.<sup>4,27</sup> Ruthenium complexes are one of the most common aqueous ECL luminophores in ECL methods<sup>35</sup> since their bright ECL emission was found by Bard and co-workers<sup>36,37</sup> in the last century. Among them, the Ru(bpy)<sub>3</sub><sup>2+</sup>/tri-*n*-propylamine (TPrA) ECL system is the sparkling focus of attention due to its dominant role in ECL *in vitro* diagnosis (IVD), but it requires potentials typically higher than 1 V. To decrease the potential imposed, we developed an n-Si/SiO<sub>x</sub>/Ni photoanode to generate efficient P-ECL of the Ru(bpy)<sub>3</sub><sup>2+</sup>/TPrA system as shown in Figure 1B. P-ECL was observed upon illumination at 810 nm at an applied potential of 0.5 V vs SCE, which is far below the common potential of Ru(bpy)<sub>3</sub><sup>2+</sup> ECL.<sup>4</sup> To improve the stability in the aqueous



**Figure 2.** (A) Schematic representation, photograph, and ECL image of the wireless minidevice. (B) Schematic diagram of the wireless chip and the ECL images with and without a diode. (C) Comparison of ECL intensities of wireless device with and without a diode. The corresponding alternating potential time profile (D) without and (E) with a diode, recorded with a digital storage oscilloscope in a parallel connection with two electrodes. Reproduced with permission from refs 1 and 2. Copyrights 2014 and 2015, respectively, American Chemical Society.

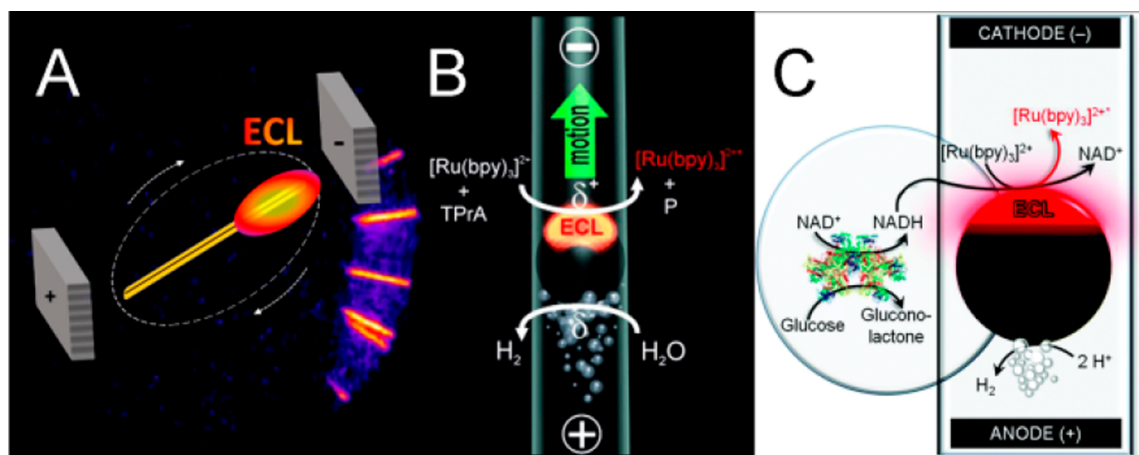
environment, the surface of Si was coated with a nickel thin film. A thin charge layer was created inside the solid at the interface between the solution and semiconductor anode which could be depleted of charge carriers depending on the applied voltage. In the n-type semiconductor photoanode system, the photo-generated holes ( $h^+$ ) were stored in the valence band and reacted with the luminophores and coreactants to accelerate their oxidation reactions and decrease the potential of P-ECL.<sup>38</sup> The reported system is a special photon upconversion process since the ECL wavelength is lower than the wavelength illuminating the semiconductor. We later designed a  $\text{BiVO}_4$  photoanode for P-ECL amplification of a luminol derivative (L-012) using illumination at 375 nm from the back side.<sup>27</sup> The incident light could simultaneously excite the depleted semiconductor anode electrode and the fluorescence of L-012. The anodic P-ECL could be generated at a low potential of  $-0.4$  V for several hours, which permitted potential applications for the biosensing and cell imaging that are vulnerable under high potential. Furthermore, Vogel et al. created a spatiotemporally controlled luminol P-ECL system by controlling the excitation light spots of 625 nm on the back side of n-type Si(111) semiconductor electrodes.<sup>38</sup> The P-ECL signals varied with the ON–OFF status and irradiation position of the excitation light. Also, the excitation from the back side could excite the luminophore under dark conditions to avoid sample damage,

photobleaching, and autofluorescence. Because of the diffusion loss of photogenerated carriers from the back side to the wet interface, it was observed that the P-ECL appeared when the thickness of the silicon electrode was below  $200 \mu\text{m}$ . As the thickness was lowered to  $70 \mu\text{m}$ , the output P-ECL pattern matched up better with the input excitation light, allowing high spatiotemporal control for P-ECL imaging. In brief, as the beneficial combination of PEC and ECL, we believe that P-ECL provides the original perspectives for the development of new detection devices and imaging strategies.

### 3. RATIONAL DESIGN OF PORTABLE DEVICES FOR ECL SENSING

#### 3.1. ECL Devices with a Wireless Energy Transfer Supply

Due to the huge demands in the development of affordable, portable, and convenient ECL instruments, different types of ECL devices keep emerging as being coupled with novel analytical technologies.<sup>22,39</sup> In a typical ECL process, the luminophore and coreactant require an electrical excitation by the external potential to emit luminescence. An electrochemical workstation is commonly used in the laboratory as an electric power source for supporting potential-resolved signals. However, its high price and inconvenience urge scientists to look for other options. For this purpose, many creative designs equipped with different electric power sources have been designed to



**Figure 3.** Schematic of ECL tracking of (A) a magnetic rotating BPE, (B) glassy carbon bead swimmers, and (C) swimmers for glucose enzymatic detection. Reproduced with permission from refs 30, 31, and 32. Copyrights 2019 American Chemical Society, 2012 Wiley-VCH, and 2014 Royal Society of Chemistry, respectively.

improve the portability and cost-effectiveness of ECL devices,<sup>22</sup> such as the primary battery,<sup>40</sup> solar cell,<sup>41</sup> supercapacitor,<sup>42</sup> and thermal power.<sup>43</sup> In addition, smartphone-based sensors are also extensively integrated because their portability and ubiquitous availability in optics, electrochemistry, and data analysis.<sup>39</sup> The universal serial bus (USB) interface and audio jack of the smartphone were also explored as the electrical stimulation source for the ECL reaction.<sup>44–46</sup>

Wireless chemical sensors have become a hot research area in recent years. Many radio technologies such as ZigBee, Bluetooth, radio frequency identification (RFID), and near-field communication (NFC) facilitate the adaption of ECL sensors in different situations.<sup>47</sup> Most of these wireless chemical sensors focus on wireless data transfer, yet NFC is one of the few that can achieve electrochemical data transfer and a power supply at the same time without a physical wire link.<sup>39</sup> Benefiting from its portable, versatile, and smartphone-compatible properties, Escobedo et al. designed a compact and battery-free NFC system that is well suited for both electrochemical and ECL detection.<sup>48</sup>

Because of the need for electrochemical stimulation to generate ECL reactions, the design of ECL devices has an analogical relationship with electrochemical devices and can use its experience for reference. Compared with electrochemical analysis, ECL detection is mainly based on the measurement of light intensity instead of current. This means that the measurement of electrochemical signals may not be necessary in ECL detection, especially for some well-investigated ECL systems. Therefore, the electric circuit of ECL devices can be simplified in many ways. Wireless power transfer (WPT) is a common wireless charging technology that was first invented by Nicola Tesla in the 1890s. The process of WPT can be described as follows. A high-frequency alternating current goes through the transmitter coil to create a time-varying magnetic field which causes the voltage in the receiver coil and achieves WPT by inductive coupling between two coils. Now the WPT technique remains active in our daily lives, such as for charging phones, cars, and human-implantable devices.<sup>49</sup> In contrast to NFC, WPT exhibits a simpler circuit with higher power, which enables a higher potential and current to support different ECL systems and multiplex ECL analysis.

In this context, we have designed several ECL devices powered by WPT.<sup>1,2,28</sup> In 2014, we first reported a WPT ECL

phenomenon with a disposable minidevice.<sup>1</sup> In this study, we tested both an electric toothbrush charger and a portable wireless charging accessory module as WPT transmitters. An enameled copper coil was connected with gold wires at both ends. The copper coil and gold wires were designed as an energy receiver and electrodes, respectively. Next, they were immersed in the carbonate buffer solution that contained luminol and H<sub>2</sub>O<sub>2</sub>. The cheap, small WPT module could be sufficiently powered by a laptop computer through a USB port for ECL reactions (Figure 2A). The output potential was easily adjusted by controlling the coil numbers of copper wire or adding a voltage regulator. When the transmitter coil and receiver coil were inductively coupled, a high frequency of alternating current (32.048 kHz) passed through the transmitter coil according to the digital storage oscilloscope monitor. Therefore, the gold wires at both ends of the copper coil were alternating between the anode and cathode at a high frequency. Two blue light spots of ECL emission were observed on these two gold wires by the naked eye and a digital camera using a relatively high concentration of luminol.

As mentioned above, the high-frequency alternating current caused the two electrodes to change alternately between the anode and cathode rapidly. As a result, luminol and hydrogen peroxide were oxidized at the positive potential to generate intermediates. Most of the generated intermediates were reduced and wasted before they participated in the ECL reaction as the potential went from positive to negative in a very short time, resulting in a weak ECL efficiency. To solve this problem, we introduced a diode into the WPT ECL system to rectify the alternating current to effectively increase the ECL intensities.<sup>2</sup> We designed a wireless electrode array chip with eight gold electrode disk-ring pairs in parallel. In the center of the chip, one coil was deposited on the front surface connected with the ring electrodes, and the other coil was deposited on the back side linked with the disk electrodes. A small, cheap diode was connected with both coils in series (Figure 2B). The diode primarily allows current to pass through in one direction and thus can be used to rectify the alternating current into a pulsating direct current (Figure 2D,E). The luminol/H<sub>2</sub>O<sub>2</sub> ECL system was used to test the feasibility of the proposed WPT system. The corresponding ECL intensity was surprisingly enhanced by 18 000-fold compared to the system without a diode (Figure 2C). We also explored the effects of different types of diodes.

Different diodes had their own maximum operating frequencies. Therefore, the diodes that had operating frequency ranges higher than the frequency of the WPT transmitter should be selected for optimal ECL enhancement. Sensitive visual  $\text{H}_2\text{O}_2$  detections were achieved by recording photographs with smartphones or digital cameras because of the dramatic increase in ECL emission.

In conclusion, the developed WPT ECL devices do not need wired connections for the power supply and have advantages such as easy manipulation, low cost, and portability. It is noteworthy that many smartphones and cheap power banks are also equipped with WPT charging functions with the recent development of WPT. A receiver with electrode arrays can also be fabricated at a low price for disposable use. Therefore, we believe that the WPT ECL devices are promising for ECL sensing.

### 3.2. Design of Bipolar Electrode Devices for ECL Sensing

A bipolar electrode (BPE) is an electric conductor that enables asymmetrical electrochemical reactions on its two poles under the control of an external electric field without a direct ohmic contact to a power supply. When a sufficiently high voltage is imposed on the driving electrodes, the resulting potential difference at both ends of the BPE will promote oxidation and reduction reactions.<sup>50–52</sup> In recent years, BPE has attracted extensive attention and has had widespread applications in chemical and biological devices because of its simplicity.<sup>21,53</sup> However, it is difficult to directly read out the electrical signals of BPE because there is no physical electrical connection between the bipolar electrode and the power supply. Fortunately, this problem was solved because the BPE was combined with the ECL readout for analysis in 2001.<sup>54,55</sup> The ease of ECL imaging enables us to simultaneously track the reactions on many BPEs, the motion of these BPEs, and the corresponding Faradaic current flowing through these BPEs.<sup>21,56</sup> For example, we have developed several ECL devices for real-time monitoring of the motion of movable BPEs, such as magnetic rotating BPEs (Figure 3A)<sup>30</sup> and glassy carbon bead swimmers (Figure 3B).<sup>31</sup> These swimmers were utilized further for the dynamic enzymatic detection of glucose (Figure 3C).<sup>32</sup> Glucose was oxidized by glucose dehydrogenase with the concomitant reduction of  $\text{NAD}^+$  to  $\text{NADH}$ , which acted as the coreactant in the ECL process. The oxidation of  $[\text{Ru}(\text{bpy})_3]^{2+}$  and the enzymatically produced  $\text{NADH}$  at the anodic pole of the BPE generated glucose-dependent ECL. Due to the wireless character of BPE, combining movable BPEs with ECL imaging provides a facile way to reveal the moving, electrochemical, and ECL properties of dynamic BPE systems.

There are two types of bipolar configurations according to the current flowing path, including closed and open BPE systems (Figure 4).<sup>51,53</sup> In a closed BPE system, two electrochemical

cells are physically separated. The BPE connected in series between two cells is the only current path, leading to a current efficiency of 100%. Compared with open BPE, the closed BPE systems usually have higher potential drops and lower current demands. Another advantage is that the anode (cathode) of the driving electrodes and the BPE anode (cathode) are separated in different cells, which can eliminate the background signal on the driving electrode to a certain degree. For example, a closed bipolar device using a modified nanopipette was developed for intracellular ECL analysis in single cells.<sup>57</sup> Pt was deposited inside the nanopipette tip, permitting intracellular molecules such as hydrogen peroxide and glucose to electrochemically transfer into the nanopipette to generate luminol ECL. Despite the advantages mentioned above, closed BPE systems are disadvantageous in terms of multiplex analysis due to the complicated fabrication. In contrast, the open BPE systems are able to readily achieve multiplex analysis using BPE arrays in the electric field formed by one pair of driving electrodes in the solution. For example, Crooks's group has developed a large-scale multiplexed open BPE microarray for ECL high-throughput analysis through simultaneous imaging of ECL emissions.<sup>58</sup> To make full use of ECL solution, we have dispersed numerous micro-/nano-BPEs (e.g., carbon microbeads, multiwalled carbon nanotubes) in solution and employed agarose gel to keep them well separated during ECL analysis. Thanks to the simultaneous wireless bipolar addressing of these BPEs, intense 3D ECL in the entire solution was generated instead of 2D ECL confined to a limited electrode surface in previous ECL devices.<sup>33</sup> This device was also able to image spatial variations of the concentration and composition of inhomogeneous samples such as a biochemical concentration gradient in a capillary configuration through one ECL image and achieve multicolor multiplex analysis.<sup>34</sup>

According to the working principle of BPE, the potential difference between two driving electrodes is larger than that of BPE. The intense ECL signals from the driving electrode may interfere with the ECL detection in open BPE systems, yet the driving electrode has rarely been studied. Recently, we developed an open BPE system with low ECL interference by the driving electrodes by using stainless steel electrodes as the driving electrodes.<sup>29</sup> As shown in Figure 5, the proposed BPE device was made on a polypropylene plate perpendicularly embedded with two stainless steel rods and two glassy carbon rods. The BPE was formed by connecting two glassy carbon electrodes with copper wire. After the polishing process, a cheap, reusable cold patch was applied for the easy construction of a reaction cell. The classic luminol/ $\text{H}_2\text{O}_2$  ECL system was used to test this BPE device. The ECL images were recorded with a smartphone in a dark box. The ECL intensities on the BPE increase as the voltage increases from 3 V and level off at 5 V. In contrast, the ECL intensities on the stainless steel driving electrode decreased from 3 V and became negligible at voltages higher than 5 V. As we mentioned above, the thin passivation layer on the stainless steel working electrode could increase the sensitivity to  $\text{H}_2\text{O}_2$  detection,<sup>26</sup> but the passivation layer became thicker as the voltage increased, which reduced the electrochemical activity and ECL intensity on the stainless steel driving electrode. The increase in ECL intensity on BPE and the decrease in ECL intensity on the stainless steel driving electrode at an appropriate high voltage demonstrated that the use of a stainless steel electrode as the driving electrode could improve the signal-to-noise ratio to enhance the sensitivity. In addition, we found that the cold patch could be more easily attached to the

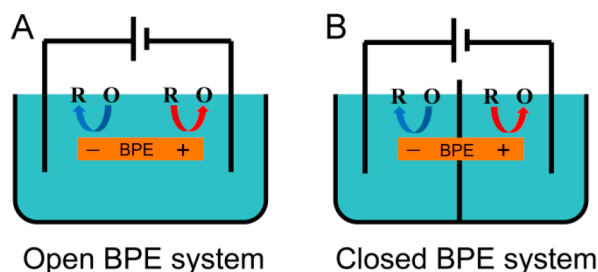
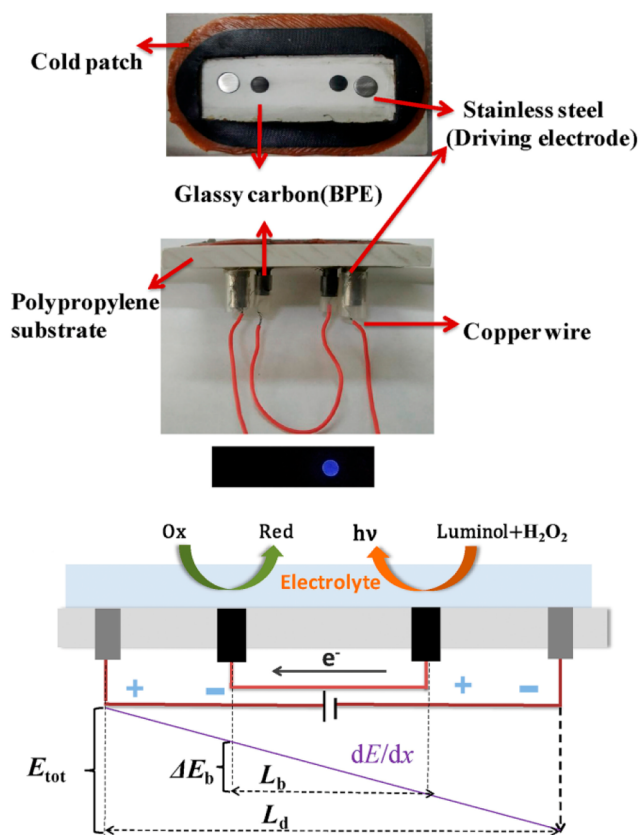


Figure 4. Schematic of (A) open and (B) closed BPE systems.



**Figure 5.** Photograph, ECL image, and schematic diagram of the regenerable BPE device. Adapted with permission from ref 29. Copyright 2017 Elsevier.

polypropylene plate repeatedly than to PDMS. The low background on the stainless steel driving electrode, repeatedly attachable cold patch, and easily polished bulk electrode make this device user-friendly for the BPE ECL study.

Besides the high interference on the driving electrodes, multiplex BPE ECL analysis requires the use of electrode arrays that are immersed in the same solution. It increases cost and makes it inconvenient to detect multiple samples. To solve these problems, we reported a single-electrode electrochemical system (SEES) using only a single film electrode with multiplex cells based on a resistance-induced potential difference.<sup>3</sup> The detailed processes are shown in Figure 6A. First, we cut the ITO glass electrode to a suitable size. Then the insulating PET label was punched to frame shape and attached to the ITO glass to build an electrochemical cell for ECL reactions. Finally, two copper wires were connected to the ITO electrode along the two edges of PET using conductive carbon ink. In the SEES, the ECL reactions were initiated through the potential difference induced by the resistance of the electrode instead of the resistance of the conductive solution in a conventional BPE system. Figure 6B,C illustrate the principle and analogous electrical circuit of the SEES. After the solution was pipetted into the cell, a voltage ( $E_{\text{tot}}$ ) was applied to both poles of the ITO electrode, and the total current ( $i_{\text{tot}}$ ) from the copper wire passed through the ITO glass ( $i_e$ ) and the solution in the cell ( $i_c$ ). We found that the SEES currents between the cells with and without the conductive solution were almost the same, indicating that the resistance of the ITO electrode ( $R_e$ ) is much smaller than the resistance of the solution ( $R_c$ ). Therefore, an approximately uniform potential gradient ( $dE/dx$ ) was formed along the ITO

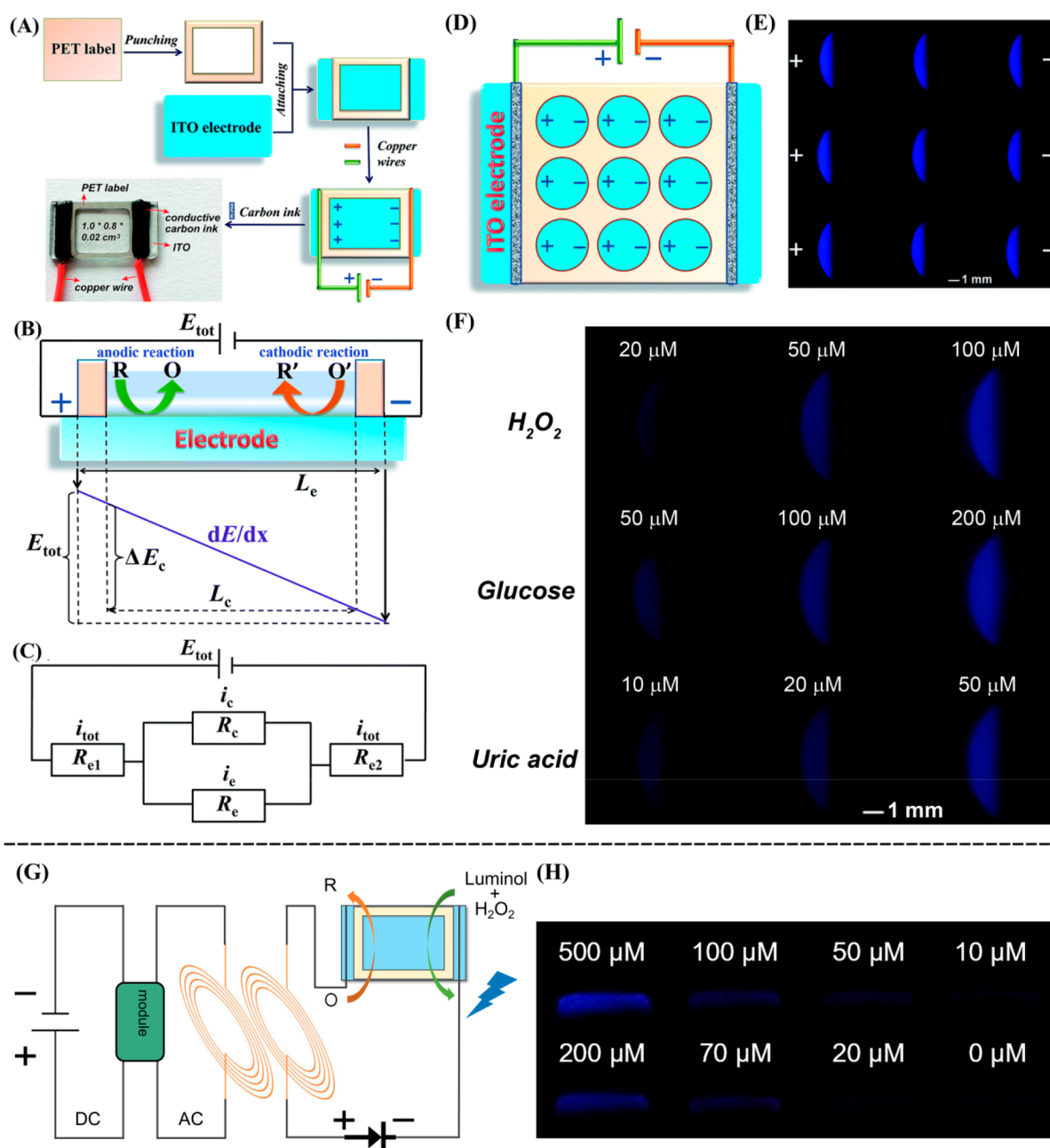
electrode based on its resistance. The potential difference ( $\Delta E_c$ ) across the cell equals the  $E_{\text{tot}}$  multiplied by the ratio of the cell length ( $L_c$ ) and the length between two copper wires ( $L_e$ ), which is illustrated by the following equation:  $\Delta E_c = E_{\text{tot}} \times L_c/L_e$ . We used the luminol/ $\text{H}_2\text{O}_2$  ECL system to verify the feasibility of SEES for ECL sensing. If the  $\Delta E_c$  was high enough,  $\text{H}_2\text{O}_2$  and dissolved oxygen were reduced at the cathode pole of the cell. In the meantime, luminol and  $\text{H}_2\text{O}_2$  were oxidized on the ITO surface at the anode pole and generated blue ECL emission. For the SEES with multiplex cells, the manufacturing processes were similar except for the simple adjustment of ITO size, the number of holes, and the arrangement in the PET according to the need. The applied potential should be enhanced responsively according to the formula to meet the  $\Delta E_c$  for ECL reactions. As shown in the Figure 6D, the nine cells of the same luminol/ $\text{H}_2\text{O}_2$  solution had nearly the same intensities with a satisfactory RSD of 2.7% (Figure 6E). Because each cell was physically isolated from others by the PET label, the solution containing different concentrations of analytes or different analytes could be simultaneously detected in this multicell SEES. Here the concentrations of  $\text{H}_2\text{O}_2$ , glucose, and uric acid were sensitively measured at the same time by analyzing the ECL image of SEES (Figure 6F). Finally, we further fabricated a SEES with 80 ( $8 \times 10$ ) cells and imposed a safe voltage of only 15 V for multiplex ECL measurements and high-throughput analysis.

To further explore the potential of SEES in the application of portable device, we developed a wireless single-electrode system by coupling SEES and the WPT technique (Figure 6G,H).<sup>28</sup> This wireless single-electrode ECL system consisted of a WPT module, a SEES, and a diode. Because the electric resistance of ITO is lower than that of solution, the SEES has relatively higher current demand compared with common two- and three-electrode systems, indicating the need for higher power for the SEES ECL device. As described above, the WPT is a highly developed technology with a wide power supply range which has been adapted for charging smartphones to electric cars. The luminol/ $\text{H}_2\text{O}_2$  ECL system was used to prove the feasibility of the integrated technique of SEES and WPT.  $\text{H}_2\text{O}_2$  was successfully detected from 1 to 150  $\mu\text{M}$  using PMT and from 10 to 100  $\mu\text{M}$  using a smartphone as a detector.

The SEES eliminates the need for the fabrication of an electrode array, avoids the interference from ECL of driving electrodes in conventional BPE ECL, and enables the simultaneous detection of multiple samples with the same voltage. Screen-printed electrodes and thin film electrodes (e.g., boron-doped diamond film electrodes) generally have some resistance and can be used as electrodes for SEES. The simplicity, versatility, and ease of the simultaneous detection of multiple samples make SEES promising for broad applications.

#### 4. SUMMARY AND OUTLOOK

ECL is one of the most popular electroanalytical methods due to its high sensitivity, easy controllability, and capability for high-throughput analysis. The ECL devices profit from the emerging multidisciplinary technology. In this Account, we summarize the rational design of ECL devices in terms of electrodes, WPT systems, and bipolar electrodes, hoping to promote the development of new ECL devices and relevant ECL applications. The P-ECL device is beneficial for exploring new light-addressable systems and their imaging applications. Stainless steel electrodes may find applications as not only working electrodes but also driving electrodes to enhance the



**Figure 6.** (A) Fabrication procedure, (B) schematic diagram, and (C) circuit diagram of the SEES. (D) SEES with nine cells and (E) corresponding ECL image of luminol/ $\text{H}_2\text{O}_2$ . (F)  $\text{H}_2\text{O}_2$ , glucose, and uric acid were detected in the multicell SEES. (G) Schematic diagram of the wireless single-electrode system. (H) Visual detection of various concentrations of  $\text{H}_2\text{O}_2$ . Reproduced with permission from refs 3 and 28. Copyrights 2018 Royal Society of Chemistry and 2019 Elsevier, respectively.

sensitivity in bipolar configurations. The boom in electronic techniques (such as WPT and NFC) can be used to build a broad platform on which wireless ECL devices can make remarkable progress. Finally, the different forms of BPE systems permit various ECL applications. For example, most screen-printed electrodes using various materials (e.g., carbon nanotubes, graphene, carbon fibers, carbon nanohorns, carbon black, carbon nitrides, MXene, two-dimensional transition-metal dichalcogenides, transition-metal dichalcogenides, perovskites, layered double hydroxides, MOF, COF, and quantum dots) have some resistance and can be fabricated at low cost. They are potential SEES electrodes for multiplex analysis, such as multiple biomarker detection and drug screening. Moreover, the dispersion of numerous BPEs in solution enables 3D ECL in

the entire solution and remarkably enhances the ECL intensity, which facilitates the development of sensitive methods using cheap, less sensitive detectors, such as smartphones. Undoubtedly, there are plenty of opportunities to design new ECL devices and related detection strategies for various applications, especially in IVD, point-of-care testing, biological studies, pharmaceutical assays, food safety, and environmental monitoring.

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