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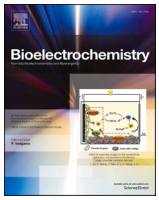

Application study of RGB color extraction in water toxicity detection

Rongbing Li [a,b], Xiaoxuan Sun [b,c], Yujia Jiang [b,d], Ling Liu [b], Yunhui Li [a,e,*], Dengbin Yu [b,d,*], Jianwei Zhu [e,*], Shaojun Dong [b,c,*]

[a] School of Chemistry and Environmental Engineering, Changchun University of Science and Technology, Changchun, Jilin 130022, PR China
[b] State Key Laboratory of Electroanalytical Chemistry, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun, Jilin 130022, PR China
[c] University of Science and Technology of China, Hefei, Anhui, 230026, PR China
[d] College of Chemistry, Jilin University, Changchun, Jilin, 130022, PR China
[e] Zhongshan Institute of Changchun University of Science and Technology, Zhongshan, Guangdong, 528437, PR China

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ABSTRACT

In this work, a colorimetric biosensor based on the rapid color change of *Escherichia coli* DH 5α (*E. coli*) and *p*-benzoquinone (BQ) was introduced to evaluate water toxicity. Here, dark brown quinhydroquinone (QHQ) was obtained from the reaction between *E. coli* and BQ. The inhibition ratios were calculated by RGB (red, green, blue) values of samples or oxidation current of hydroquinone (HQ). Here, RGB technology was used for biotoxicity assessment for the first time. The results suggested that the biosensor-based RGB has an obvious dose-dependent response. The IC_{20} values of Hg^{2+} and Cu^{2+} obtained based on RGB values were 1.0 and 1.5 mg L^{-1}, respectively. This biosensor can be used to detect quickly toxicity of the Cu^{2+} and Hg^{2+} in water without complicated operation steps. Therefore, the biosensor has potential application value in the field of water toxicity detection.

1. Introduction

With rapid economic development, extensive use of chemicals has produced a lot of pollutants that are discharged into the environment as a result of human activity. Pollutants, including heavy metal ions and organic matters in water, not only cause tremendous harm to the environment but also threaten the health of humans. Therefore, it is of great practical importance to develop new methods of water pollution detection for the evaluation of biological toxicity. Water quality detection methods include physical and chemical methods (high-performance liquid chromatography (HPLC) [1,2], inductively coupled plasma-mass spectrometry (ICP–MS) [3–5], and ultraviolet–visible (UV–vis) [6,7]) as well as biological methods. Conventional physical and chemical analysis methods can analyze quantitatively the types and concentrations of toxic substances in water, and possess a low limit of detection. However, the disadvantages of these methods are that the synergy or antagonism of different metal ions [8] cannot be detected, and these approaches require complicated pretreatment and detection steps.

Traditional biological toxicity detection methods are mostly based on biological endpoint methods, such as mortality, growth ratio, and reproduction ratio, which are relatively effective water monitoring methods. Fish [9], Daphnia [10,11], algae [12] and higher animals [13,14] are used as indicating organisms to evaluate the comprehensive toxicity of wastewater by detecting the inhibitory effects of toxic substances on the movement, growth and development as well as respiratory activity of indicator organisms. However, these methods are usually complicated, time-consuming and expensive in practical applications, and real samples require preprocessing or cleaning before analysis [12,15]. Therefore, it is necessary to adopt a reliable and rapid approach for biotoxicity evaluation. Microorganisms have received widespread attention due to their high sensitivity, low cost, rapid response, and convenience of storage and so on. In addition, microbial sensors have also been employed in some key fields such as environmental monitoring [16–18] and food quality management [19]. Toxicity detection methods based on microorganisms in water mainly include luminous bacteria technique [20,21], mediated method [22,23], nitrifying

Abbreviations: *p*-benzoquinone, BQ.

* Corresponding authors at: Zhongshan Institute of Changchun University of Science and Technology, Zhongshan, Guangdong, 528437, PR China (Y. Li, J. Zhu) and State Key Laboratory of Electroanalytical Chemistry, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun, Jilin 130022, PR China (D. Yu, S. Dong).

E-mail addresses: liyh@cust.edu.cn (Y. Li), yudbs@ciac.ac.cn (D. Yu), zhujw@jlu.edu.cn (J. Zhu), dongsj@ciac.ac.cn (S. Dong).

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bacteria method [24] and bioelectrochemical method [25–27]. The electrochemical mediated method has certain advantages in that test is not interfered with by chromaticity or turbidity. Among these methods, electrochemical/colorimetric dual signal detection methods have attracted much attention owing to the addition of visual signals. However, on account of the limitations of sensitivity in the naked eye colorimetric detection and differences in the human body, limit of detection of the colorimetric method is not low enough. With scientific and technological advances, digital products such as digital cameras, smartphones [28] and scanners are becoming increasingly popular in digital imaging colorimetry [29,30]. These digital products can sometimes play a vital role in scientific research. In particular, camera-equipped smartphones have been gaining popularity among scientific researchers in recent years. Analysis methods based on RGB [31–34] (red, green, blue) detection with smartphones have also received increasing attention. However, there is no report on the application of RGB technology in the field of water toxicity detection.

As a lipophilic redox mediator, *p*-benzoquinone (BQ) can enter the cell to accept electrons from the electron transport chain. It is applied widely to evaluate the toxicity of water in the construction of biosensors. In this work, a water toxicity detection method based on the rapid color change between *Escherichia coli* DH 5α (*E. coli*) and BQ was established. *E. coli* was utilized as the model bacteria, and BQ [35] was used as the electron mediator. RGB values obtained through smartphone and color software were used first to detect the toxicity of wastewater containing heavy metal ions or organic compounds. On the basis of feasibility verification, test conditions containing cell concentration, detection time and detection range were optimized by the RGB method. The toxicity of wastewater containing Cu^{2+} or Hg^{2+} was detected sensitively under optimal conditions. It is worth mentioning that the experiment was less harmful to the operators because the experimental system was closed. Therefore, the results showed that the novel method was simple, rapid, safe and intuitive. This method has important application prospects in the development of colorimetric toxicity detection equipment in the future.

2. Materials and methods

2.1. Reagents and chemicals

Peptone was purchased from Beijing Aoboxing Biotechnology Co., ltd.. Yeast extract was purchased from British Oxoid Co., ltd. Glycerol was acquired from Beijing Ding Guo Changsheng Biological Technology Co., ltd.. NaCl, KCl, NH_4Cl, $NaH_2PO_4{\cdot}2H_2O$, and $Na_2HPO_4{\cdot}12H_2O$ were obtained from Beijing Chemical Factory. $Pb(CH_3COO)_2{\cdot}3H_2O$ was purchased from Beijing Hongxing Chemical Factory. $HgCl_2$ was purchased from Jiangyan Huanqiu Reagent Factory. $Cu(CH_3COO)_2{\cdot}H_2O$ and BQ were acquired from Sinopharm Group Chemical Reagent Co., ltd.. $ZnSO_4{\cdot}7H_2O$ was acquired from Beijing Pinggu Shuangyan Chemical Plant. $Co(NO_3)_2{\cdot}6H_2O$ was purchased from Xilong Science Co., ltd.. $Ni(CH_3COO)_2{\cdot}3H_2O$ was obtained from Tianjin East China Reagent Factory. 3,5-dichlorophenol (3,5-DCP) and *p*-nitrophenol (PNP) were purchased from Sigma-Aldrich and Shanghai General Chemical Reagent Factory, respectively. Luria broth (LB) was prepared with 10% peptone, 5% yeast extract, and 10% NaCl, and the pH was adjusted to 7.4 with 1 M NaOH. Phosphate buffer solution (PBS, 0.2 mM) was prepared with 0.08 M Na_2HPO_4 and 0.12 M KH_2PO_4[36]. Several solutions containing heavy metal ions of Cu^{2+}, Hg^{2+}, Pb^{2+}, Zn^{2+}, Ni^{2+} and Co^{2+} were diluted to the desired concentrations from 1000 mg L^{-1}. 0.625 mM BQ was prepared with 50 mM PBS, and then it was wrapped with aluminum foil and stored in a 4 °C refrigerator. Unless otherwise stated, all solutions were prepared with deionized water (≥18.2 MΩ/cm) purified by a Milli-Q Plus system (Millipore).

2.2. Apparatus

E. coli was incubated in an Oscillator (ZHWY-200B, Shanghai Zhicheng Analytical Instrument Manufacturing Co., ltd.). UV−vis−NIR spectrometer (Varian) was utilized to detect the optical density (OD_{600}) of *E. coli* at 600 nm. Centrifuge (5804 R, Eppendorf Co. ltd.) was used to separate culture medium and *E. coli*. The current–time curves were obtained on an electrochemical workstation (CHI 832B, Shanghai Chen Hua Instrument Co., ltd.) by a conventional three-electrode system with an as-prepared Pt microarray electrode (MAE) (50.0 μm diameter Pt wire, 32 pieces) [37] as the working electrode, Ag/AgCl electrode as the reference electrode and Pt wire as the auxiliary electrode. Here, the MAE was polished with a 0.05 μm γ-alumina slurry and sonicated in deionized water before use.

2.3. Microbial culture

E. coli was purchased from the China Microbial Culture Collection and Management Center (CGMCC). LB was sterilized at 121 °C in an autoclave for 20 min. The purified *E. coli* suspended in LB containing 30% glycerol was used as strains and stored in a −80 °C refrigerator. *E. coli* was inoculated in LB and cultivated for 14 h at 37 °C using a shaker at 200 rpm. *E. coli* was centrifuged for 7 min at 5000 rpm at room temperature, and then the cells were washed and centrifuged twice with 1 % NaCl [38] solution to remove the LB. Finally, *E. coli* was resuspended in 1% NaCl solution and stored in a 4 °C refrigerator for subsequent experiments. Cells were only used on the day of collection.

2.4. Principle of toxicity detection

Scheme 1 illustrates a diagram of water toxicity detection based on RGB analysis. BQ is reduced to hydroquinone (HQ) by electrons released by *E. coli* respiration, and then red brown QHQ is obtained by the further reaction between the remaining BQ and HQ. The respiratory activity of *E. coli* is inhibited when toxic substances exist in the sample. Less QHQ is obtained because the amount of reduced BQ decreases, which causes the sample color to become lighter. A smartphone (Mi 10) was utilized to take pictures, and RGB values were read by MATLAB 7.01. Finally, the inhibition ratios were calculated with RGB values of r/(r + g + b).

For the colorimetric method, the inhibition ratio is obtained by the ratio of RGB values of r/(r + g + b) according to Eq. (1).

$$Inhibition\% = \left(1 - \frac{(r/(r+g+b))_{Tox}}{(r/(r+g+b))_{Con}}\right) \times 100\% \quad (1)$$

Here, r, g and b are the values of the three primary colors of light, $(r/(r+g+b))_{Tox}$ is the ratio of the RGB values of the toxic sample, and $(r/(r+g+b))_{Con}$ is the ratio of the RGB values of the control sample.

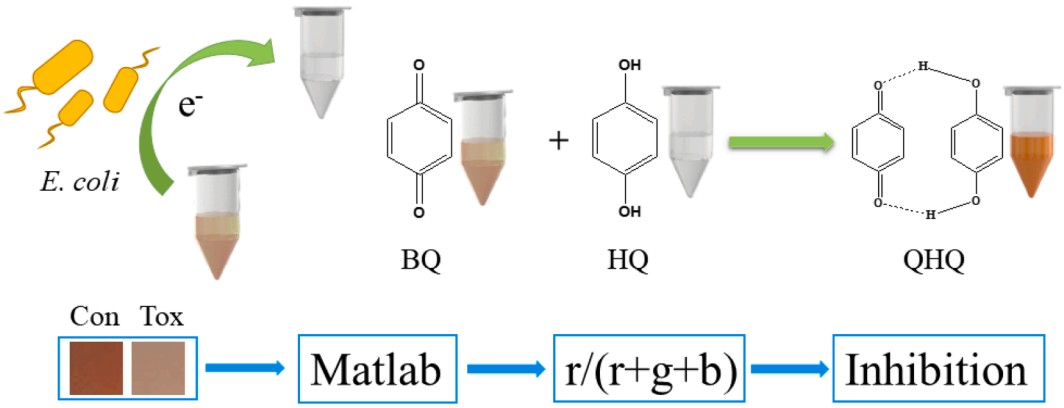
For the electrochemical method, the inhibition ratio is obtained by the current values according to Eq. (2).

$$Inhibition\% = \left(1 - \frac{I_{Tox}}{I_{Con}}\right) \times 100\% \quad (2)$$

I_{Tox} is the steady-phase current of the sample to be tested in the presence of toxic substances, and I_{Con} is the steady-phase current of the control sample in the absence of toxic substances.

2.5. Toxicity detection procedure

The two-step method is more accurate than the one-step method in detecting the toxicity of wastewater containing heavy metal ions by the mediator method [39]. Therefore, in this study, the two-step method was adopted to prevent producing precipitation of the reaction between heavy metal ions and phosphate of PBS and BQ, which caused the actual concentrations of heavy metal ions to be lower than the expected results [40].



Scheme 1. Diagram of BQ-mediated biotoxicity detection based on RGB analysis.

Artificial wastewater containing Cu^{2+}, Hg^{2+}, Ni^{2+}, Pb^{2+}, Zn^{2+}, Co^{2+}, PNP or 3,5-DCP was employed as a toxic substance. A total volume of 1 mL of the incubation suspension was injected into a 1.5 mL centrifuge tube for each sample. The incubation suspension contained 0.9 mL of cell suspension with the desired concentration and 0.1 mL of certain toxic substances with various concentrations as the sample to be tested. Here, the toxic substance was replaced with deionized water of the same volume for the control sample. The toxins were mixed first with *E. coli* cells, and then incubated for some time. All samples were washed to clear out the toxins by centrifuging twice at 11000 rpm for 7 min with 1% NaCl solution. Then, 1 mL 0.625 mM BQ [41] was added to the harvested cells. After a suitable reaction time, the colors of the samples could be visually distinguished, and photographs of the samples were taken with a smartphone under fixed light. Segments of the same area were cut from the original photographs of the samples, in which RGB values were read by MATLAB software. After 1 h, all supernatants were obtained by centrifuging samples at 11000 rpm for 7 min, and then the current values of the supernatants were measured by chronoamperometry. Unless otherwise stated, all experiments were performed three times in parallel, and all colorimetric and electrochemical experiments were performed at room temperature.

3. Results and discussion

3.1. Feasibility of toxicity detection

The feasibility verification of toxicity detection is shown in Fig. 1. The *E. coli* suspension was diluted to $OD_{600} = 2.4$ with 1% NaCl solution. The photograph was shot by a smartphone after mixing *E. coli* and BQ for 3 min. Segments of the same area taken from the same position of the three sample photographs are shown in Fig. 1A. The BQ sample was very light brown, and the control sample showed a red brown. However, the color of the toxic sample was light brown, which indicated that the amount of produced QHQ diminished in the toxic sample. The current–time curves of the control samples, toxic samples and BQ samples are shown in Fig. 1B. The current of the samples without Cu^{2+} was higher than that of the samples with Cu^{2+}, which indicated that the respiration of *E. coli* was inhibited. We could clearly see the difference between them from Fig. 1C. The above results showed that there were consistent detection results by both colorimetric and electrochemical methods, which can distinguish toxic and nontoxic water samples. Therefore, it was feasible to develop a colorimetric toxicity biosensor using the rapid color reaction between *E. coli* and BQ.

3.2. Optimization of bacterial concentration

The effect of bacterial concentration on detection sensitivity is shown in Fig. 2. The images in Fig. 2 were taken from the photographs of samples after 5 min (Fig. S1). The inhibition ratios were calculated according to the RGB values of each photograph in Fig. 2. Fig. 2 shows that the constructed RGB toxicity evaluation system exhibited a dose-dependent response. More electrons were released as increase of bacterial concentration. The inhibition ratios increased from 3.53% to 41.87% with increasing bacterial concentration in the range of OD_{600} = 0.3–1.2. Subsequently, the inhibition ratios decreased with increasing bacterial concentration in the range of OD_{600} = 1.2–4.8. The inhibition ratio and detection sensitivity were the highest at the cell concentration of OD_{600} = 1.2. Therefore, OD_{600} = 1.2 was chosen as the optimal cell

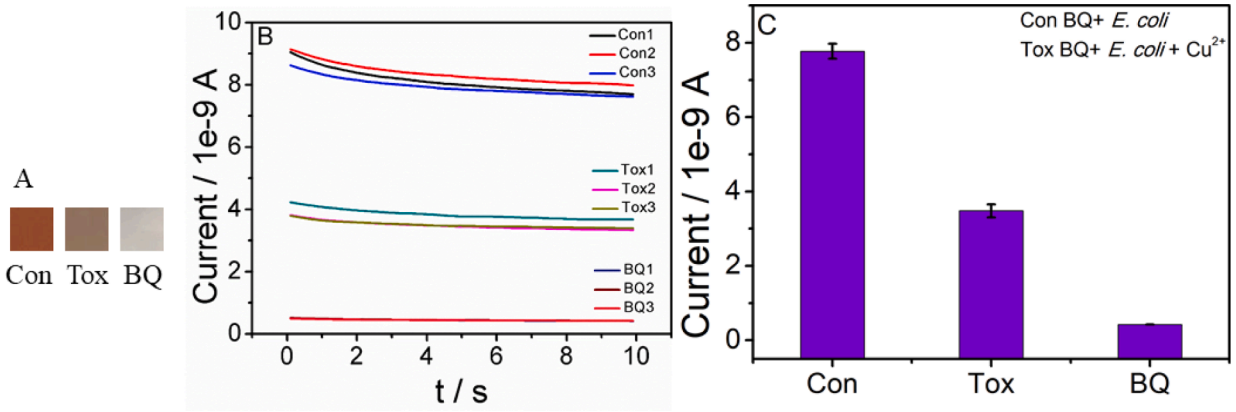


Fig. 1. (A) Photographs of three kinds of samples of Con1 – Con3 (*E. coli* + BQ), Tox1 – Tox3 (*E. coli* + Cu^{2+} + BQ) and BQ1 – BQ3 (BQ) at 3 min. Concentrations of *E. coli*, BQ, and Cu^{2+} were OD_{600} = 2.4, 0.625 mM and 4 mg L^{-1}, respectively; (B) Current-time curves of the corresponding samples in (A); (C) Current bar chart of the corresponding samples in (A).

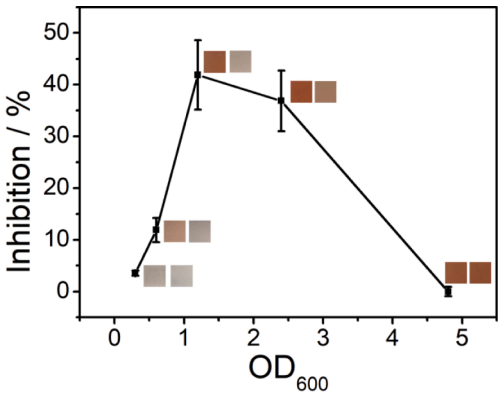


Fig. 2. The effect of concentrations of *E. coli* on the biotoxicity assessment of Cu^{2+} (OD_{600} of *E. coli* were 0.3, 0.6, 1.2, 2.4 and 4.8). 0.625 mM BQ and *E. coli* reacted with 4 mg L^{-1} Cu^{2+} for 1 h at room temperature, and photographs were taken from samples after 5 min of reaction.

concentration for subsequent experiments, which was adequate to obtain a distinct response.

3.3. Optimization of reaction time

The optimization of the reaction time is shown in Fig. 3. Photographs were obtained from samples at 1, 2, 3, 4, 5, 6, 8 and 10 min (Fig. S2), respectively. As shown in Fig. 3, compared with the colors of the control samples, the colors of the poisoned samples were lighter, which indicated that the amount of QHQ of the toxic samples was less than that of the control samples. The inhibition ratio was very small, and the colors of the control and toxic samples were lighter at 1 min. This is because both the nonpoisoned bacteria and the poisoned bacteria produced less QHQ obtained from the reaction of HQ and unreacted BQ. The inhibition ratios increased with the extension of reaction time until 3 min. This may be caused by the rapid production of QHQ in control samples and the slow production of QHQ in toxic samples. Over time, the production of HQ and QHQ further reduced due to the decrease in bacterial activity in the toxic samples, so the inhibition ratios were further increased. At 4–6 min, the inhibition ratios tended to be stable, and the inhibition ratio reached a maximum of 41.2% at 6 min, these results indicated that the amount of QHQ produced in the control samples and in the toxic samples tended to be stable with increasing time. At 6–10 min, the inhibition ratios decreased slightly, which was also related to the different reaction ratios between nonpoisoned bacteria, poisoned bacteria and BQ. This can be attributed to the following reasons. The reaction between the nonpoisoned bacteria and BQ was steady, while the amount of HQ obtained by the reaction between the poisoned bacteria and BQ still increased slowly. As a result, the difference in the produced QHQ between the two reactions decreased, so the inhibition ratios decreased slowly. Therefore, 6 min was chosen as the optimal reaction time in subsequent experiments.

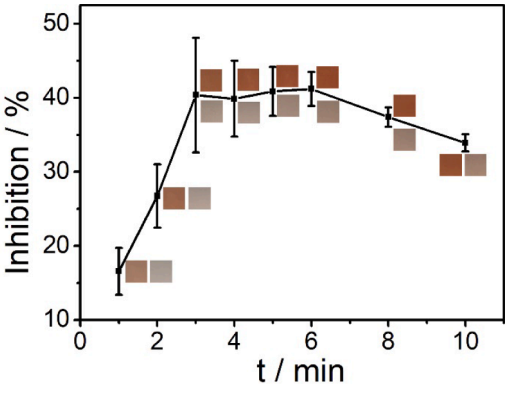


Fig. 3. Effect of reaction time on the inhibition ratio of toxicity detection. OD_{600} = 1.2 *E. coli*, 0.625 mM BQ with 4 mg L^{-1} Cu^{2+} reacted for 1, 2, 3, 5, 6, 8 or 10 min at room temperature.

3.4. Response sensitivity of the biosensor to different toxic substances

The sensitivity of a biosensor for toxic compounds is an essential factor that should be considered. Thus, under the optimal conditions, the responses of *E. coli* to six heavy metal ions (Pb^{2+}, Ni^{2+}, Zn^{2+}, Co^{2+}, Hg^{2+} and Cu^{2+}) and two phenolic compounds (PNP and 3,5-DCP) were analyzed according to the RGB values of the samples. The photographs in Fig. 4A were taken from the photographs of each sample after 6 min of reaction (Fig. S3). It is obvious from Fig. 4A that the colors of the samples including Cu^{2+} and Hg^{2+} were lighter than that of the control sample. However, the colors of samples containing other toxic substances were similar to those of the control sample. This indicated that the amount of QHQ was significantly lower in the two samples than in the other samples. That is, microbial activity was lower, and the amount of HQ obtained was also lower.

As shown in Fig. 4B, the inhibition ratios of Hg^{2+} and Cu^{2+} were 21.5% and 16.4% respectively. Other heavy metal ions and phenolic compounds showed low inhibition or no inhibition to *E. coli*. Obviously, the inhibition ratios of the two toxic substances were significantly higher than those of the other toxic substances at the same concentrations. These results indicated that the two toxic substances possessed high sensitivity to *E. coli* in this system. Therefore, the colorimetric biosensor is more suitable for detecting wastewater containing Cu^{2+} or Hg^{2+}.

3.5. Toxicity detection of wastewater

Under the optimal conditions, the toxicities of wastewater containing Hg^{2+} or Cu^{2+} were detected according to RGB values and current values. The final concentrations of both Hg^{2+} and Cu^{2+} were 0.05, 0.1, 0.2, 0.4, 0.8, 1.6, 3.2, 6.3 and 12.5 mg L^{-1}, respectively. Pictures taken from each sample photograph are shown in Fig. 5A and 5B, and inhibition ratio curves are shown clearly in Fig. 5C and 5D.

The RGB values decreased as the Hg^{2+} and Cu^{2+} concentrations increased, and the developed RGB toxicity evaluation system exhibited a dose-dependent response. It can be observed from Fig. 5A that the colors of the samples of Hg^{2+} lightened gradually in the range of 0.4–3.2 mg L^{-1} and almost no change in the range of 3.2–12.5 mg L^{-1}. As shown in Fig. 5C, there were low inhibition ratios, and the inhibition ratios declined slowly when the concentration was 0.4–3.2 mg L^{-1}. This is because the stress response of *E. coli* led to an increase in *E. coli* activity, and the amount of produced QHQ increased. The inhibition ratios increased slowly when the stress response began to decrease or even disappeared with the continuous increase in the Hg^{2+} concentration. When the concentration of Hg^{2+} was 0.4–3.2 mg L^{-1}, the inhibition ratios increased continuously. This result suggested that the activity of *E. coli* and the amount of produced QHQ decreased due to the increase in Hg^{2+} concentration. In the range of 3.2–12.5 mg L^{-1}, the inhibition ratios tended to be stable. It was easy to understand that almost all cells were poisoned, which led to the loss of cells reactivity, and no QHQ was produced at higher concentrations of Hg^{2+}, so the inhibition ratios were almost constant.

In Fig. 5B, the colors of the samples of Cu^{2+} faded gradually from red brown to light brown in the range of 0.4–3.2 mg L^{-1} and were almost unchanged in the range of 3.2–12.5 mg L^{-1} as the Cu^{2+} concentration increased. From the image in Fig. 5D, the inhibition ratios continued to increase when the concentration of Cu^{2+} was 0.05–3.2 mg L^{-1}, which indicated that these were not stress reactions for low concentrations of Cu^{2+} compared to Hg^{2+} of the same concentrations. The activity of *E. coli* decreased with increasing concentrations of Cu^{2+}, and the amount of BQ reduced to HQ decreased, which resulted in the amount of

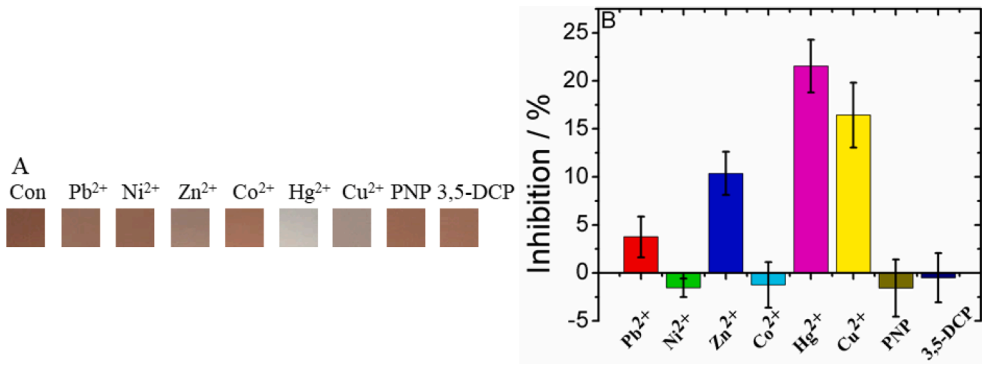


Fig. 4. Response sensitivity of the biosensor to different toxic substances. (A) Screenshots of smartphone photographs were taken from samples containing 0.625 mM BQ and OD_{600} = 1.2 *E. coli* after reacting for 6 min with 10 mg L^{-1} different toxins; (B) Histograms of inhibition ratios obtained from RGB values of the corresponding photographs in (A).

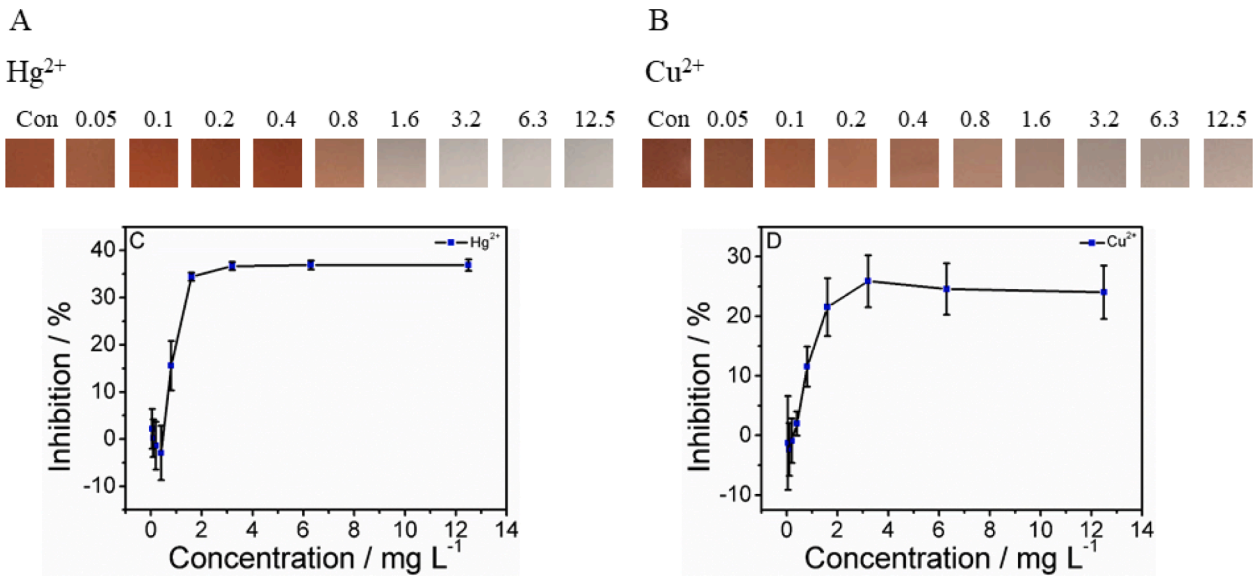


Fig. 5. Screenshots of toxicity test samples contained 0.625 mM BQ, *E. coli* of OD_{600} = 1.2 reacted for 6 min at room temperature of (A) Hg^{2+} and (B) Cu^{2+}; The inhibition ratio curves of Hg^{2+} (C) and Cu^{2+} (D) at different concentrations at 6 min.

produced QHQ decreased, thus, the inhibition ratios increased. When the concentration of Cu^{2+} was 3.2–12.5 mg L^{-1}, the inhibition ratios were almost changed. The reason was the same as that for Hg^{2+}, as almost all cells were poisoned and inactivated, and no more QHQ was produced at higher concentrations of Cu^{2+}.

As shown in Fig. 5C and 5D, the IC_{20} values of both Hg^{2+} and Cu^{2+} were 1.0 and 1.5 mg L^{-1}, respectively, which were deduced from the inhibitory curves. The amount of HQ in the supernatant solutions was detected by chronoamperometry (0.3 V vs Ag/AgCl). The inhibition ratios were obtained from the current values (Fig. S4), which can be used as supplementary tests. The trends of the inhibition ratio curves by the electrochemical method were identical to those of the RGB method. In addition, a comparison of IC_{50} values obtained by the electrochemical method is shown in Table S1. The linear calibration functions for Hg^{2+} and Cu^{2+} are shown in Fig. S5, and the parameters of the linear equation obtained from Fig. S5 was shown in Table S2. Meanwhile, the method proposed was compared with other methods reported in Table S3.

4. Conclusions

In this study, a biosensor combining the RGB method and the electrochemical method was designed to detect toxicity in water. The color reaction between BQ and *E. coli* was further studied by popular smartphone. RGB values of the photographs were read through MATLAB software, and then the toxicity inhibition ratios were calculated. The IC_{20} values of Hg^{2+} and Cu^{2+} obtained by the colorimetric method were 1.0 and 1.5 mg L^{-1}, respectively. Here, the RGB method was used first to evaluate water toxicity. In summary, this study provided a rapid, novel, visual and safe method without complex detection procedures and large instruments in water toxicity detection. It has certain application prospects in the detection of wastewater containing Cu^{2+} or Hg^{2+}.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.bioelechem.2022.108270.

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