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A Low-Cost Colorimetric Detection of Hg^{2+} using Paper-based Sensor with Euclidean Distance Approach

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Abstract

Mercury (Hg) is a highly toxic heavy metal that poses significant risks to human health and aquatic ecosystems. The development of rapid and low-cost detection methods is urgently needed. In this work, the paper-based colorimetric sensor was developed by immobilizing dithizone onto Whatman No. 41 filter paper for the detection of Hg^{2+}. The sensor utilized the color change of the sensor from green to orange with the color formation of Hg–dithizone complex. FTIR analysis confirmed the successful immobilization of dithizone with characteristic absorption of C=N and C=S vibrations. For quantification, digital images captured by a smartphone were analyzed using the Euclidean Distance (ED) approach. The sensor demonstrated a linear response in the concentration range of 1–5 mg/L (R^2 = 0.991), with a limit of detection (LOD) and limit of quantification (LOQ) of 0.50 mg/L and 1.67 mg/L, respectively. Furthermore, the sensor exhibited high precision with a %RSD of 4.39%. Selectivity tests under acidic conditions confirmed that Hg^{2+} yielded the highest ED value compared to other metal ions such as Cu^{2+}, Pb^{2+}, Co^{2+}, Ni^{2+}, and Zn^{2+}. This paper-based sensor provides a simple, portable, and cost-effective tool for the on-site monitoring of mercury in environmental samples

Keywords: mercury, paper-based sensor, colorimetric

Abstrak

Mercury (Hg) merupakan logam berat yang sangat beracun dan menimbulkan resiko serius bagi kesehatan manusia dan ekosistem perairan. Pengembangan metode deteksi yang cepat dan murah saat ini sangat dibutuhkan. Pada penelitian ini, sensor kolorimetri berbasis kertas telah dikembangkan dengan mengimobilisasi ditizon pada kertas saring Whatman No. 41 untuk mendeteksi Hg^{2+}. Sensor ini memanfaatkan perubahan warna dari hijau ke orange akibat pembentukan kompleks ditizon-Hg. Analisis FTIR telah mengkonfirmasi keberhasilan imobilisasi ditizon dengan penyerapan karakteristik gugus C=N dan C=S. Untuk kuantifikasi, gambar digital diperoleh dengan smartphone dan dianalisis menggunakan pendekatan Euclidean Distance (ED). Sensor menunjukkan respons linier pada rentang konsentrasi 1–5 mg/L (R^2 = 0.991), dengan batas deteksi (LOD) dan batas kuantifikasi (LOQ) masing-masing 0.50 mg/L dan 1.67 mg/L. Selain itu, sensor menunjukkan presisi dengan %RSD sebesar 4,39%. Uji selektivitas pada kondisi asam menunjukkan bahwa Hg^{2+} menghasilkan nilai ED tertinggi dibandingkan dengan ion logam lain seperti Cu^{2+}, Pb^{2+}, Co^{2+}, Ni^{2+}, dan Zn^{2+}. Sensor berbasis kertas ini dapat menjadi alat yang sederhana, portabel, dan hemat biaya untuk pemantauan merkuri secara langsung pada sampel lingkungan.

Kata kunci: merkuri, sensor berbasis kertas, kolorimetri

1. Introduction

Mercury (Hg) is a highly toxic heavy metal because it can chemically transform in aquatic environments into more dangerous forms, such as methylmercury, which is neurotoxic and easily bioaccumulated (Hakim *et al.*, 2025). Mercury exposure can cause serious disorders of the central nervous system, kidney damage, cognitive impairment, and developmental abnormalities in children and fetuses (Yan *et al.*, 2014; Zargoosh and Farhadian Babadi, 2015). In aquatic ecosystems, mercury accumulates in aquatic organisms and undergoes biomagnification throughout the food chain, increasing the risk of exposure to humans through the consumption of fish and aquatic biota. Therefore, the presence of mercury in water, even at low concentrations, remains a serious problem to human health and the sustainability of aquatic environments. The primary sources of mercury pollution are mostly from human activities such as gold mining, cosmetic waste, industrial waste, fossil fuel combustion, etc (Hakim and Bilad, 2024). For drinking water, the World Health Organization (WHO) has set the regulation for Hg^{2+} at 6 µg/L(Nshnsh *et al.*, 2023), while in Indonesia, according to Minister of Health Regulation No. 2 of 2023, the maximum Hg^{2+} level is 1 µg/L (Anonim, 2023). Therefore, the accurate, rapid, and inexpensive detection methods for Hg^{2+} have been a challenge in research over the past few decades.

Currently, several selective and sensitive methods for Hg^{2+} analysis have been applied, such as CV-AAS (Lins *et al.*, 2019), ICP-OES (Friedrich *et al.*, 2024), ICP-MS (Chen *et al.*, 2025), (AFS) (da Silva Cunha *et al.*, 2022), AES (Rong *et al.*, 2022), and electrochemistry (Ganesh *et al.*, 2025). However, these methods are relatively expensive, require complex preparation, and cannot be used in in-situ analysis. The method currently being developed is colorimetry. The colorimetric method provides simpler, easy to use, rapid, and on-site application. The colorimetric method will produce a color change in the matrix that indicates the presence of an analyte, including heavy metal ions (Chen *et al.*, 2023). The intensity of color change can be assessed visually, using a smartphone, or spectrophotometry, which can be directly correlated with the concentration of an analyte.

Several studies have reported the utilization of smartphones in the analysis of metal ions using colorimetry methods. Reported by (Preechaburana, Sangnuy and Amloy, 2023) successfully detected Hg^{2+} using a smartphone on a filter paper matrix modified with urea-capped AuNPs. Shrivas et al. (Shrivas *et al.*, 2020) successfully developed a sensor paper modified with AgNPs/CTAB and captured it with a smartphone camera. Attaallah and Amine (Attaallah and Amine, 2022) reported the use of smartphones in detecting Cd^{2+} in drinking water using horseradish peroxidase enzyme inhibition. In this work, the Euclidean Distance approach was used to analyze the RGB values generated by the sensor. The analysis was conducted on the conditions before and after the sensor came into contact with the sample solution.

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2. Methodology

2.1. Materials and instruments

The materials utilized in this research included ethanol p.a., dithizone, mercuric chloride ($HgCl_2$), nitric acid (HNO_3), zinc sulfate ($ZnSO_4$), nickel (II) sulfate ($NiSO_4$), copper (II) sulfate ($CuSO_4$), lead (II) nitrate ($Pb(NO_3)_2$), cobalt nitrate ($Co(NO_3)_2$), Whatman filter paper No. 41, and distilled water.
FTIR spectroscopy (Shimadzu IR Prestige-21 spectrometer) was used to analyze the functional group. The absorbance of the solution was measured using UV-Vis Spectrophotometer (Safas Monaco SP2000). The photography process was carried out using a smartphone camera (Redmi 9C, 13 MP).

2.2. Preparation of the paper-based sensor

The filter paper used was Whatman Filter Paper No. 41, which has a porosity and thickness of 25 µm and 215 µm, respectively. Filter paper was cut into 2x10 cm pieces, then dipped into a dithizone solution (0.05% in ethanol solvent) and dried at room temperature.

2.3. Preparation of the paper-based sensor

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To analyze the sensor's performance, the paper-based sensors were contacted with the analyte by dipping the sensor into Hg^{2+} solutions (acidic conditions). The resulting colors were then captured using a smartphone camera. The RGB values were extracted using ImageJ software and converted into ED values using the following equation (Hakim and Bilad, 2024).

$$ED = \sqrt{(\Delta R)^2 + (\Delta G)^2 + (\Delta B)^2}$$

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3. Results and Discussion

3.1. Characterization of the paper-based sensor

The paper-based sensor has been successfully developed with a green color on its surface, which is characteristic of dithizone. The sensor produces a color change after interacting with Hg^{2+} solution and forms an orange color. This color is produced from a complex reaction between dithizone and mercury. Figure 1b shows the absorbance of the sensor due to the formation of a complex compound between Hg^{2+} metal ions and dithizone in the visible region. This absorption is possible due to electron transitions in the d orbital and charge transfer in the complex compound, which requires relatively few energy in the visible region. The maximum absorbance of the Hg^{2+}-dithizone complex was obtained at a maximum wavelength of 488 nm, as reported in the study by Juliana et al., (Juliana, Kurniaty and Miftah, 2016). The maximum wavelength is slightly different from the maximum wavelength of the Hg-dithizone complex, which is 490 nm in micelle media (Khan, Ahmed and Bhanger, 2005), 495 nm in chloroform solvent (Zaetun *et al.*, 2015).

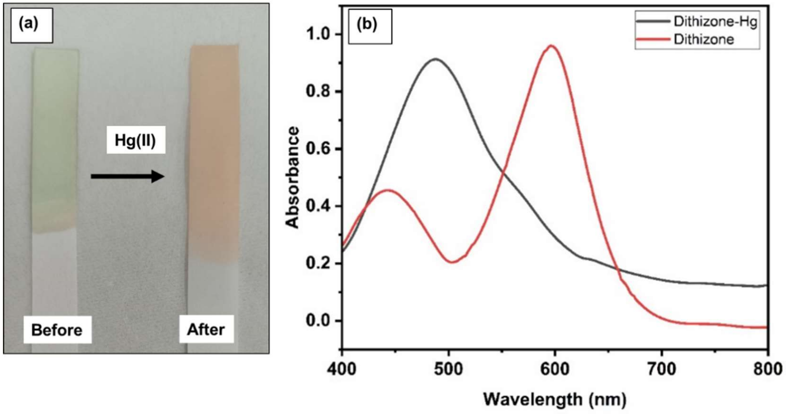

Figure 1. Visualization of the paper-based sensor before and after reacting with Hg^{2+} solution (a) and UV-Vis spectra of free dithizone and dithizone-Hg complex

FTIR characterization was performed to confirm the successful immobilization of dithizone on the Whatman filter paper matrix by comparing the shift in wave numbers on specific functional groups. Based on Figure 2a, which is pure dithizone, an absorption peak was detected at a wave number of 3330 cm^{-1}, which is the stretching vibration of the N-H group. The aromatic and functional structure of dithizone was confirmed by the appearance of C=N strain absorption at 1647 cm^{-1}, as well as the characteristic vibration of the C=S group, which appeared at 1087 cm^{-1} and 1046 cm^{-1}. Additionally, N-H bending vibrations and aromatic ring stretching vibrations are observed in the regions of 1455 cm^{-1} and 1379.66 cm^{-1}. Figure 2b shows a spectral pattern that is a combination of the characteristics of dithizone and the cellulose matrix of Whatman filter paper. The appearance of broad and strong peaks in the range of 3332 cm^{-1} to 3277 cm^{-1} indicates an overlap between the N-H stretching vibrations of dithizone and the hydroxyl (O-H) groups that are very dominant in the cellulose structure. According to (Nur, Rohaeti and Darusman, 2017) the presence of NH groups overlapping with OH group absorption. The presence of the filter paper matrix is confirmed by the appearance of a sharp absorption peak at 1029 cm^{-1}, which is the C-O vibration of cellulose fibers (Hospodarova, Singovszka and Stevulova, 2018), as well as absorption at 2898 cm^{-1}, which originates from the aliphatic C-H stretching of cellulose. The presence of the C=N absorption peak at 1646 cm^{-1} indicates the successful immobilization of dithizone in the pores of the filter paper. The slight shift in the C=N peak from 1647 cm^{-1} to 1646 cm^{-1} indicates intermolecular interactions, possibly in the form of hydrogen bonds or physical entrapment of dithizone molecules within the cellulose fiber network.

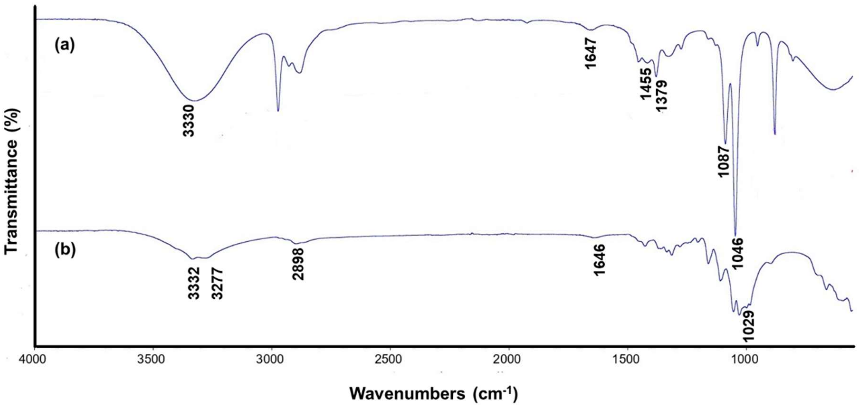

Figure 2. The IR spectra of dithizone (a) and filter paper immobilized by dithizone

3.2. Linearity, limit of detection (LOD) and limit of quantification (LOQ)

Linearity is the ability of the sensor to provide a proportional response to the concentration of analytes in a sample. Linearity has a working range, which is the lowest and highest limits of analytes that can be determined accurately and precisely. The limit of detection (LOD) and limit of quantification (LOQ) were calculated by determining the slope of the calibration curve and the standard deviation from triplicate measurements. The LOD indicates the lowest concentration of analyte that produces a response from the sensor, and the LOQ indicates the minimum concentration that can be quantified reliably and with acceptable precision (Neera, Nidhi and Ureena, 2020). In this work, the range of concentration obtained was between 1-5 mg/L. A calibration curve was established between Euclidean Distance (ED) value and Hg^{2+} concentration (Figure 3) with a linear equation of y=19.11x + 67.56 and R^2= 0.991, while the LOD and LOQ values were 0.50 and 1.67 mg/L, respectively.

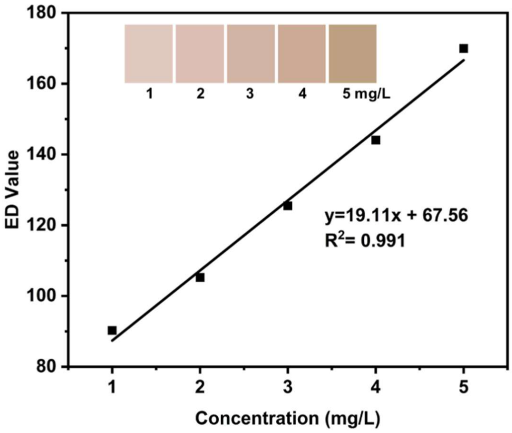

Figure 3. Calibration curve of the paper-based sensor

3.3. The effect of interference metals

The study of metal ion interference has been conducted. The paper-based sensor was dipped into a metal ion solution, and the resulting color was captured with a smartphone. The results showed differences in the intensity of the colors produced by the formation of dithizone complexes with metal ions. Figure 4 shows the effect of various metal ions on the ED value under acidic conditions. The ED value quantifies the colorimetric response in the sensor due to the formation of metal-dithizone complexes, where a higher value indicates a stronger interaction. The Hg^{2+} ion shows the highest ED value, indicating the most stable complex formation and the most significant color change. This is related to the high affinity of Hg^{2+} for sulfur and nitrogen donor atoms in the dithizone structure, resulting in a very stable and intensely colored complex even under acidic conditions. Cu^{2+} ions exhibit relatively high ED values but lower than Hg^{2+}.

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Pb^{2+} and Co^{2+} ions show intermediate and relatively similar ED values, indicating a moderate sensor response to both ions. Acidic conditions are thought to cause partial protonation of the active group of dithizone, thereby reducing its coordination effectiveness with these ions. Ni^{2+} ions give higher ED values than Zn^{2+}, but still lower than Hg^{2+} and Cu^{2+}, indicating the formation of complexes with limited color intensity. Meanwhile, Zn^{2+} shows the lowest ED value, indicating the weakest interaction and minimal color change.

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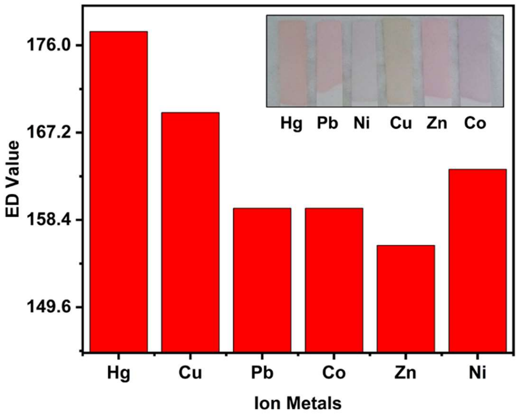

Figure 4. Response of the sensor with other metals in ED value

3.4 Precision

Precision indicates the consistency of the sensor in producing the same measurement values repeatedly when measuring the same object or condition and usually expressed in %RSD. Precision values of the proposed sensor were analyzed by extracting RBG values and converting them to ED values for different types of sensors (n=10). The %RSD obtained was 4.39%.

4. Conclusions

This study successfully developed a low-cost and portable paper-based colorimetric sensor for Hg^{2+} detection using dithizone as a chromogenic agent. The immobilization of dithizone on the filter paper matrix was confirmed through FTIR spectroscopy, which shows significant intermolecular interactions between the ligand and cellulose fibers. The sensor showed a distinct color change from green to orange upon interaction with Hg^{2+}, allowing for quantitative analysis via a smartphone-based Euclidean Distance approach. Analytical results indicated a strong linear correlation (R^2 = 0.991) within the 1–5 mg/L range, an LOD of 0.50 mg/L, and repeatability with %RSD of 4.39%. Selectivity studies demonstrated that the sensor is highly responsive to Hg^{2+} even in the presence of interfering ions, which is attributed to the high affinity of Hg^{2+} for the sulfur and nitrogen donor atoms in dithizone. This proposed sensor has great potential for rapid, in situ mercury monitoring in water resources.

Acknowledgement

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